GDR Groupement de recherche
UP Ultrafast Phenomena

Réunion plénière du GDR UP - 2023

11-13 décembre 2023

Cité internationale universitaire de Paris

Recueil des contributions

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

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

Cette septième réunion plénière du GDR qui se tient du 11 au 13 décembre 2023 à la Cité Internationale Universitaire de Paris est une occasion importante de rapprochement entre les équipes françaises et une démonstration de l'émulation scientifique qui caractérise notre communauté. En plus des sessions habituelles, cette année la réunion plénière est complétée par des interventions sur le thème "Grandes infrastructures internationales de recherche pour les sciences ultrarapides" avec des représentants de Eu-XFEL, ELI-ALPS, ELI-Beamlines qui présenteront leurs installations et les opportunités associées pour notre communauté, notamment en lien avec les sciences attosecondes.

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

Le Bureau du GDR UP :

Franck Lépine (ILM, Lyon),
Lionel Poisson (ISMO, Saclay),
Fabrice Catoire (CELIA, Bordeaux),
Pascal Salières (LIDYL, Saclay),
Morgane Vacher (CEISAM, Nantes),
Jérémie Caillat (LCPMR, Paris),
Benjamin Lasorne (ICGM, Montpellier),
Pascale Changenet (LOB, Palaiseau),
Vincent De Waele (LASIR, Lille),
Tatiana Itina (LaHC, St Etienne),
Christine Richter (LPMS, Cergy),
Pascal Ruello (IMMM, Le Mans),
Jérôme Faure (LOA, Palaiseau),
Sophie Kazamias (IJCLab, Orsay),
Damien Bigourd (IMS, Bordeaux),
Sébastien Weber (CEMES, Toulouse),
Fabien Vialla (ILM, Lyon),
Sarah Houver (MPQ, Paris).

Lundi 11 décembre 2023
Salon Honnorat

09h00-10h00	***ACCUEIL***	
10h00-10h30	**BUREAU DU GDR UP**	Introduction Actions 2023 du GDR, Actions prévues, Opportunités
SESSION 1: Attoscience (Chair: Sophie Kazamias)		
10h30-11h10	***Subhendu Kahaly***	***ELI - ALPS***
11h10-11-30	Eric Mevel CELIA, Bordeaux	XUV high order harmonic wavefront and mode control
11h30-11h50	Juliette Dubois LOA, Palaiseau	Frequency resolved cross correlation between XUV high harmonics and IR fundamental laser pulses by transient multiphoton absorption spectroscopy in gases
11h50-12h10	Verónica Oliver Álvarez de Lara ULTRAFAST innovation	From classic to state-of-the-art instruments to generate, characterize, and manipulate ultrafast light
12h10-13h40	***REPAS / EXPOSANTS***	
SESSION 2: High intensity XUV & Xray (Chair: Marc Simon)		
13h40-14h20	***Sakura Pascarelli***	***Eu-XFEL***
14h20-14h40	Saikat Nandi iLM, Lyon	Generation of quantum entanglement between two massive particles using a seeded free-electron laser
14h40-15h00	Maurizio Monti IMMM, Le Mans	Surface melting of orbital order in an ultrafast phase transition
15h00-15h20	Weipeng Yao LULI, Palaiseau	Characterization of proton and X-ray generation at the Apollon short-focal-area in the 1-2 PW range
15h20-15h50	***PAUSE CAFÉ / EXPOSANTS***	
SESSION 3: High Intensity IR (Chair: Annie Klisnick)		
15h50-16h30	***Jakob Andreasson***	**ELI-Beamlines**
16h30-16h50	Aline Vernier LOA, Palaiseau	Laplace HC : un accélérateur laser-plasma conçu pour les applications
16h50-17h10	Adrien Kraych IJCLab, Orsay	Slowing down the light in vacuum with intense laser pulses
17h10-17h30	Francois Sylla SOURCELAB	KAIO-Beamline – a modular high-repetition rate laser-plasma electron accelerator for a broad range of applications
	Photo conference	
17h30-19h00	***Session Poster***	
17h30-20h30	***Cocktail***	

Mardi 12 décembre 2023
Salon Honnorat

09h00-09h30	***Accueil Café***	
SESSION 4 : Condensed phase (Chair: Pascale Changenet)		
09h30-09h50	Emmanuel Benichou iLM, Lyon	Probing the organization of liquids from the bulk to the interface by second harmonic generation
09h50-10h10	Manuel Llansola-Portoles I2BC, Saclay	Regulation of excited states in photosynthetic systems
10h10-10h30	Chloé Magne ISMO, Orsay	Ultrafast spectroscopy to explore Multiple Exciton Generation
10H30-11h00	***PAUSE CAFÉ / EXPOSANTS***	
SESSION 5: Materials 1 (Chair: Christine Richter)		
11h00-11h20	Siham Benhabib LPS, Orsay	Non-adiabatic Lifshitz transition in High Tc superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$
11h20-11h40	Yannis Laplace LSI, Palaiseau	Ultrasmall and tunable TeraHertz surface plasmon cavities at the ultimate plasmonic
11h40-12h00	Clément Hainaut PhLAM, Lille	Ultrafast measurement of turbulent phenomena in a quantum fluid-of-light
12h00-12h20	Niloufar Nilforoushan LMPQ, Paris	Intense and ultrabroadband THz sources at a 200 kHz repetition rate for time-resolved nonlinear study of low-energy excitations
12h20-14h00	***REPAS / EXPOSANTS***	
SESSION 6: Isolated systems 1 (Chair: Victor Despré)		
14h00-14h20	Alexandra Viel IPR, Rennes	Ultra-fast or ultra-slow dynamics through conical intersections? The case of NO_3
14h20-14h40	Constant Schouder ISMO, Orsay	Solvation dynamics of an alkali ion in a helium nanodroplet
14h40-15h00	Lina Fransén CEISAM, Nantes	Ultrafast photochemistry of ionized ethylene $C_2H_4^+$: a theoretical perspective
15h00-15h20	Joachim Galiana ICGM, Montpellier	Quantum dynamics around PPE'S conical intersections for spectroscopic and real-time studies
15h20-15h50	***Pause café / EXPOSANTS***	
SESSION 7: Materials 2 (Chair: Angela Vella)		
15h50-16h10	Mauro Fanciulli LPMS, Cergy	Ultrafast Hidden Spin Polarization Dynamics of Bright and Dark Excitons in 2H-WSe_2
16h10-16h30	Laurène Gatuingt LMPQ, Paris	Out-of-equilibrium dynamics in the Bi2212 antiferromagnetic phase
16h30-16h50	Jelena Sjakste LSI, Palaiseau	Electron-phonon coupling and ultrafast dynamics of hot carriers in semiconductors: from interpretation of photoemission experiments to transport simulations in devices.
16h50-17h10	Paolo Maioli iLM, Lyon	Ultrafast spectroscopy investigations of heat transfer at the nanoscale
AFTERWORK		

Mercredi 13 décembre 2023
Salle Gulbenkian

09h00-09h30	***Accueil Café***	
SESSION 8: Secondary sources 1 (Chair: Jean-Christophe Delagnes)		
09h30-09h50	Pierre Béjot ICB, Dijon	PI-FROSt: “Plasma-Induced Frequency Resolved Optical Switching”
09h50-10h10	Slava Smartsev LOA, Palaiseau	Simple few-shot method for spectrally resolving the wavefront of an ultrashort laser pulse
10h10-10h30	Francesco Massimo LPGP, Palaiseau	Recent progress in the modeling of laser wakefield acceleration
10h30-11h00	***Pause Café / discussions***	
SESSION 9: Isolated systems 2 (Chair : Valérie Veniard)		
11h00-11h20	Emmanuel Fromager LCQ, Unistra	Ensemble density functional theory of electrons and nuclei
11h20-11h40	Rafael Menezes Ferreira LIDYL, Saclay	Probing two-photon resonant ionization with electron interferometry
11h40-12h00	Anthony Ferté CEISAM, Nantes	Quantum interference effects in the post-ionization dynamics of fluoro-benzene unravelled through non-adiabatic simulations
12h00 – 13h30	***REPAS***	
SESSION 10: Isolated systems 3 (Chair: Morgane Vacher)		
13h30-13h50	Morgan Berkane LCPMR, Paris	Theoretical study of electron-nuclei entanglement in molecular photoionization
13h50-14h10	Francesco Talotta LPCT, Nancy	How to tackle the radiationless transitions via quantum chemistry methods
14h10-14h30	Raman Maksimenka FASTLITE-Amplitude	Towards table-top and high-flux OPCPA drivers at 2.1 µm for Soft-X-Ray generation.
14h30-15h00	***Pause Café / discussions***	
SESSION 11: Secondary sources 2 (Chair: Stefan Haessler)		
15h00-15h20	Igor Andriyash LOA, Palaiseau	Circumventing limits of Laser-Plasma Acceleration with advanced optics
15h20-15h40	Stylianos Passalidis LMCE, Bruyères-le-Chatel	Modelling electron deflectometry measurements of magnetic fields in ultrahigh-intensity, femtosecond laser-foil interactions
15h40-16h00	Luca Fedeli LIDYL, Saclay	Probing strong-field QED with Doppler-boosted ultra-intense lasers
16h00-16h15	***Conclusions***	

Programme des afterworks mardi 12 décembre

Afterwork jeunes (organisé par le Club Jeunes du GDR UP) :

Rendez-vous à partir de 17h30 au bar :

« Le Corvisart »
43 Boulevard Auguste Blanqui
75013 Paris 13

Afterwork permanents :

Rendez-vous à partir de 17h30 :
« Cuccagna »
69 Rue de la Glacière
75013 Paris 13

https://www.cuccagna.fr/

COHERENT

OPTON LASER INTERNATIONAL

SESSION 1

Attoscience

Subhendu Kahaly
ELI – ALPS

XUV high order harmonic wavefront and mode control

E. Mével[1], Sylvain Prawdziak[1], C. Picot[2], B. Miller[7], C. Valentin[1], K. Veyrinas[1], M. Plach[3], J. Peschel[3], M. Hoflund[3], F. Catoire[1], P. Smorenburg[4], H. Dacasa[3], S. Maclot[3], C. Guo[3], H. Wikmark[3], V. Strelkov[5,6], C. Arnold[3], P. Eng-Johnsson[3], A. Zaïr[7], A. L'Huillier[3], E. Constant[2]

1. Centre Lasers Intenses et Applications (CELIA), Université de Bordeaux-CNRS-CEA, 33405 Talence, France
2. Université Claude Bernard Lyon 1, CNRS, Institut Lumière Matière, 10 rue Ada Byron, 69622 Villeurbanne
3. Department of Physics, Lund University, SE-221 00 Lund, Sweden
4. ASML Research, ASML Netherlands B.V., 5504 DR Veldhoven, The Netherlands
5. Prokhorov General Physics Institute of the Russian Academy of Sciences, 38, Vavilova Street, Moscow 119991, Russia.
6. Moscow Institute of Physics and Technology (State University), 141700 Dolgoprudny, Moscow region Russia
7. King's College London, Department of Physics, Attosecond Physics Laboratory, Strand WC2R 2LS London, United Kingdom

It is well known, especially since October 2023, that high order harmonics generated in gases produces attosecond pulses that can be used to track ultrafast electrons dynamics. A current and a very active field is to use the brightness and the coherence of harmonic source to develop applications in high resolution imaging and spatial metrology. Both types of applications required focusing a broadband XUV source on a target. However, it was shown that the different harmonics are focused at places that are longitudinally separated [1 - 6] thus limiting the intensity and the spatial homogeneity of the attosecond structure.

Here, we study the XUV harmonics spatial mode and wavefront at the sources and when refocused. We use the SWORD [2] method that provides such a 1D characterization for several harmonic orders. We compare the XUV beam properties when generated with a truncated Gaussian beam or a flat top driving laser beam. We observe that Flat-top spatial shaping reduces the XUV chromatic aberrations [7] as compared to the case of using a truncated Gaussian beam. Intermediate configurations with super-Gaussian diverging or converging driving beams are under current studies to combine chromatic aberrations mitigation, large enough generating medium and micro XUV sources.

References:

[1] "Spatiotemporal structure of the combined field of high-order harmonics and generation of attosecond pulses", Platonenko V.T., Strelkov V.V, Quantum Electronics, **27**, 779 (1997).

[2] "Order dependent structure of high harmonic wavefront, E. Frumker", G. G. Paulus, H. Niikura, A. Naumov, D. M. Villeneuve, and P. B. Corkum, Opt. Expr. **20**, 13870 (2012).

[3] "Imaging the source of high harmonics generated in atomic gas media", S. Chatziathanasiou, S. Kahaly, D. Charalambidis, P. Tzallas, and E. Skantzakis, Opt. Expr. **27**, 9733 (2019).

[4] "Focusing properties of high-order harmonics", Maria Hoflund, *et al.*. Ultrafast Science, 9797453 (2021).

[5] "Optics-less focusing of high-order harmonics", L. Quintard, V. Strelkov, J. Vabek, O. Hort, A. Dubrouil, D. Descamps, F. Burgy, C. Péjot, E. Mével, F. Catoire, and E. Constant, Science Advances **5**, eaau7175 (2019).

[6] "Spatio temporal coupling of attosecond pulses", H. Wikmark, C. Guo, J. Vogelsang et al. proceedings of the National academy of sciences, **11** 4779 (2019).

[7] "Chromatic aberrations correction of attosecond high-order harmonic beams by flat-top spatial shaping of the fundamental beam", K. Veyrinas, M. Plach, J. Peschel, M. Hoflund, F. Catoire, C. Valentin, P. Smorenburg, H. Dacasa, S. Maclot, C. Guo, H. Wikmark, A. Zaïr, V. Strelkov, C. Picot, C. Arnold, P. Eng-Johnsson, A. L'Huillier, E. Mével, E. Constant, New J. Phys. **25**, 023017 (2023)

Frequency resolved cross correlation between XUV high harmonics and IR fundamental laser pulses by transient multiphoton absorption spectroscopy in gases

Juliette Dubois[1,2,*], Léonardo Rico[2], Julien Gautier[1], Fabien Tissandier[1], Boris Vodungbo[1], Camille Lévêque[1], Jérémie Caillat[2], Richard Taïeb[2], and Guillaume Lambert[1]

[1]LOA, Ensta Paris, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, 828 Bd des Maréchaux, 91762 Palaiseau, France
[2]Sorbonne Université, CNRS, Laboratoire de Chimie Physique - Matière et Rayonnement, LCPMR, 75005, Paris, France

*juliette.dubois.loa@ensta-paris.fr

Finding a way to measure ultrashort events has led to the development of femtosecond duration pulses. For the last decades, with the development of the IR (Infra-Red) femtosecond laser, numbers of techniques have emerged in order to measure these specific pulses [1,2] . We present here a new technique to measure the temporal properties of any XUV sources from picosecond to femtosecond pulse duration. This technique is based on a cross-correlation between an IR pump pulse and an XUV probe pulse. In our particular case, the XUV probe pulse is composed of high harmonics generated from gases. The correlation is made through the interaction of the pump and the probe with a second gas. We observed that the absorption can change in a large spectral area for energies below ionization levels of gases when the IR and the XUV overlap and are synchronised (insert of Fig.1).

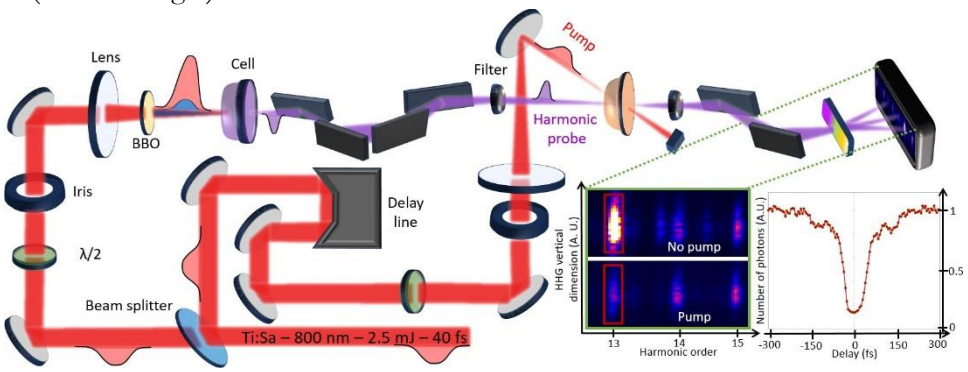


Figure 1: Top view of the IR pump-XUV probe experimental setup - Down left : spectrometer image of probe harmonics on a CCD camera, showing a series of harmonics measured after passing through 25 mbar Ne gas cell and with or without the presence of a800 nm pump beam - Down right : vertical integration of the signal of the 13th harmonic in function of the delay

Our explanation is that this phenomenon is linked to a multiphoton transient absorption involving one XUV photon and several IR photons. The absorption phenomenon therefore results from a coherent superposition of the IR and XUV pulses. Thereby, this interaction should provide temporal information on the longest pulse between the probe and the pump. In order to verify this hypothesis, we notably introduced a 3 cm long fused silica cube in the laser path of the pump in order to expand the pump pulse duration from 40 fs to 450 fs FWHM. In this case, the pump pulse duration is much longer than the harmonics one, estimated around 10 fs. As expected, we measured an absorption phenomenon very close to 450 fs. In addition, the cube was inserted in the laser path of the probe. Harmonics pulse duration was estimated around 135 fs, larger than the 40 fs of the pump pulses. Here, the absorption phenomenon was measured about 140 fs.

With our technique, it becomes possible to obtain a cross correlation trace, which provides the temporal intensity profile and the pulse duration of any XUV sources, as long as these probe pulses are longer than pump pulses. Moreover, these measurements have been made all together with spectra, giving in theory access to phase information using iterative phase-retrieval algorithms [3]. We could be able to fully reconstruct XUV pulses from various picosecond down to a few femtoseconds timescale facilities. This would be vital for applications and especially for the ones using high harmonics and for which the phenomena, to be studied, can be resonant, based on different types of energy edges or related to energy levels to be probed in a system.

References

[1] B. Wang et al., Optics Express, **24**, 22210, (2016).
[2] D.J. Kane and R. Trebino, IEEEJ. Quantum Electron., **29**, 571–579, (1993).
[3] Nils C. Geib et al., Optica, **6**, 495, (2019).

From classic to state-of-the-art instruments to generate, characterize, and manipulate ultrafast light

Verónica Oliver Álvarez de Lara

UltraFast Innovations GmbH, Dieselstr. 5, 85748 Garching b. München, Germany

oliver@ultrafast-innovations.com

Since 2009, UltraFast Innovations (UFI®) develops and manufactures a wide range of specialized and/orcustom-made optics and optical components, as well as optical devices for ultra-short pulse laser applications spanning from the IR to the XUV/soft X-ray region. Through detailed descriptions and examples of their applications, this talk will cover optical devices relevant for short-pulse generation andcharacterization, and XUV science.

SESSION 2

High intensity XUV & Xray

Sakura Pascarelli

Eu-XFEL

Generation of quantum entanglement between two massive particles using a seeded free-electron laser

S. Nandi[1], A. Stenquist[2], A. Papoulia[2], E. Olofsson[2], L. Badano[3], M. Bertolino[2], D. Busto[2], C. Callegari[3], S. Carlström[2], M. B. Danailov[3], P. V. Demekhin[4], M. Di Fraia[3], R. Feifel[5], G. Gallician[6], L. Giannessi[3], M. Gisselbrecht[2], P. Johnsson[2], M. Manfredda[3], M. Meyer[7], C. Miron[6], J. Peschel[2], O. Plekan[3], K. C. Prince[3], R. J. Squibb[5], M. Zangrando[3], F. Zapata[2], S. Zhong[2], J. M. Dahlström[2]

[1]Université Lyon, Université Claude Bernard Lyon 1, CNRS, Institut Lumière Matière, F-69622,Villeurbanne, France
[2]Department of Physics, Lund University, SE-22100 Lund, Sweden
[3]Elettra-Sincrotrone Trieste, Basovizza, 34149 Basovizza, Trieste, Italy [4]Institute of Physics and CINSaT, University of Kassel, 34132 Kassel, Germany [5]Department of Physics, University of Gothenburg, 41258 Gothenburg, Sweden
[6]Université Paris-Saclay, CEA, CNRS, LIDYL, 91191 Gif-sur-Yvette, France
[7]European XFEL, 22869 Schenefeld, Germany

saikat.nandi@univ-lyon1.fr

Entanglement is at the center of the ongoing second quantum revolution [1], but producing entanglement between macroscopic systems is a challenging task. One of the reasons is thought to be the experimental impossibility to entangle certain degrees of freedom that can permeate to the everyday classical world [2]. Here we address this issue by considering a new type of bipartite system composed of a photoelectron and a light-dressed atomic ion. We employ intense extreme ultraviolet (XUV) pulses from FERMI [3], a seeded free-electron laser, to generate and study the ultrafast development of entanglement between these two particles. We demonstrate both experimentally and theoretically that the high intensity of a femtosecond XUV pulse from FERMI can be used to generate quantum entanglement between two massive objects [4]. By adjusting the intensity of the laser pulse, we can control the manifestation of the entanglement. Our findings willbe valuable for the community as improving light sources allow not only observing but also tocharacterize and eventually take advantage of entanglement in these ultrafast processes at short wavelengths.

References:

[1] Aspect, A. The Second Quantum Revolution: From Basic Concepts to Quantum Technologies,Chapter 2, in *Photonic Quantum Technologies: Science and Applications* (2023), edited by Benyoucef, M.

[2] Jost, J. D. *et al.*, Nature **459**, 683-685 (2009).

[3] Allaria, E. *et al.*, Nature Photonics **6**, 699 (2012).

[4] Nandi, S. *et al.*, submitted (2023).

Surface melting of orbital order in an ultrafast phase transition

Maurizio Monti[1, 2], Khalid M. Siddiqui[2], Daniel Perez-Salinas[2], Xiang Li[3], Danylo Babich[4], Xin Liu[4], Mathias Sander[4], Henrik Till Lemke[4], Roman Mankowsky[4], Xuerong Liu[3], Simon Wall[2]

1. Institue des Molécules et Matériaux du Mans, UMR CNRS 6283, Avenue Olivier Messiaen 72085, Le Mans Cedex 09, France
2. Institute of physics and astronomy, Aarhus university, Ny Munkegade 120, Aarhus, 8000, Denmark
3. ShanghaiTech, 393 Middle Huaxia Road, Pudong, Shanghai, 201210, China
4. SwissFEL, Paul Scherrer Institut, Forschungsstrasse 111, Villigen, 5232, Switzerland

When studying photo-induced phase transitions, an element that is often neglected is the role played by the surface. Heterogeneity at the surface plays a key role in equilibrium phase transitions, but its role outside of equilibrium is unknown. Generally, at equilibrium, the surface melts at a lower temperature than the bulk, which can give rise to new properties of the system. This is also true for quantum materials, where electronic phase transitions (such as orbital ordering) have shown significant surface melting effects [1]. To address this, we used surface-sensitive x-ray scattering, performed at the Bernina beamline (SwissFEL), to probe the melting of the orbital order (OO) of a $La_{0.5}Sr_{1.5}MnO_4$ (LSMO) single crystal. LSMO has previously shown strong surface melting effects in equilibrium [1, 2], and its photo-induced phase transition is well studied out of equilibrium [3]. We measured time-resolved reciprocal space mapping of the orbital truncation rod (OTR, see panels a and b in the figure) and the orbital Bragg peak (OBP). Studying the OBP and the OTR we find evidence of a different threshold for the suppression of surface and bulk order, indicating surface melting on an ultrafast time scale. Moreover, studying the OTR we find an increase of diffuse scattering in large regions of the Brillouin zone, together with vibrational coherence (instead strongly suppressed in the Bragg peak), which we attribute to coherent motion of spectator atoms (panel c in the figure).
These observations point in the direction of a transition mediated by disorder, rather than a coherent one.

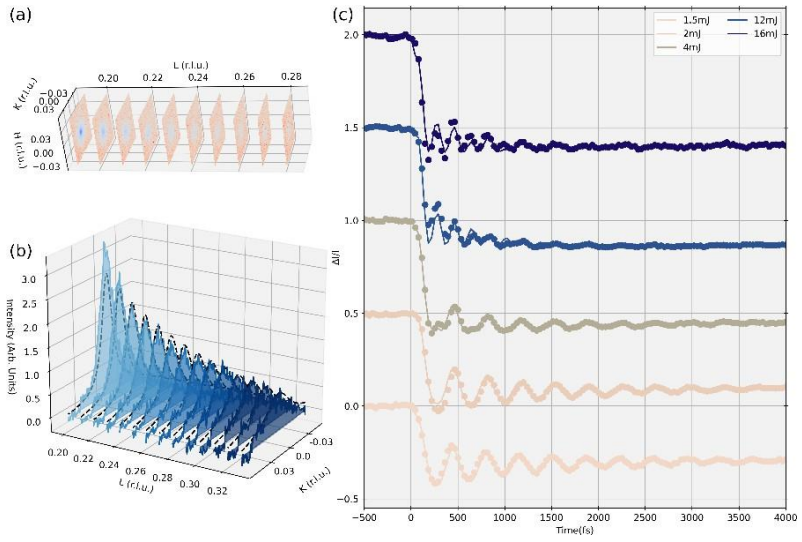


Figure: (a) reciprocal space map of the photoinduced intensity change of the OTR at 1ps, the intensity of the OTR itself decreases upon photoexcitation, while an increase of diffuse scattering appears. (b) Static profile of the OTR summed along the H direction (blue lines) with global fit of the rod intensity (dashed black line). (c) Dynamics of the intensity of the OTR as a function of excitation fluence, the presence of coherent phonons is evident. The traces have been displaced vertically for clarity

References:

[1] Y. Wakabayashi, M. Upton, S. Grenier, *et al.*, Surface effects on the orbital order in the single-layered manganite La0.5Sr1.5MnO4, Nat. Mat., **6**, 972-976, (2007)
[2] S.B. Wilkins, X. Liu, Y. Wakabayashi, *et al.*, Surface melting of electronic order in La0.5Sr1.5MnO4, Phys. Rev. B, **84**, 165103 (2011)
[3] D. Perez-Salinas, A.S. Johnson, D. Prabhakaran, *et al.*, Multi-mode excitation drives disorder during an ultrafast melting of a C4-symmetry-broken phase transition, Nat. Comm., 13, 238 (2022)

Characterization of proton and X-ray generation at the Apollon short-focal-area in the 1-2 PW range

W. Yao[1,2], R. Lelièvre[1], T. Waltenspiel[1], I. Cohen[3], A. Beck[4], E. Cohen[4], D. Michaeli[4], I. Pomerantz[3], C. Gautier[5], D. Papadopoulos[1], F. Mathieu[1], F. Trompier[6], Q. Ducasse[7], A. Allaoua[7], L. Gremillet[8,9], E. d'Humières[10], and J. Fuchs[1]

1. Laboratoire pour l'Utilisation des Lasers Intenses, UMR 7605 CNRS-CEA-École Polytechnique-Université Paris VI, 91128 Palaiseau, France
2. Sorbonne Université, Observatoire de Paris, Université PSL, CNRS, LERMA, F-75005, Paris, France
3. The School of Physics and Astronomy, Tel Aviv University, Tel-Aviv, 6997801, Israel
4. Physics department, NRCN, PO Box 9001, Beer-Sheva, Israel
5. LANL, PO Box 1663, Los Alamos, New Mexico 87545, USA
6. Laboratoire de dosimétrie des rayonnements ionisants, PSE-Santé/SDOS, IRSN, 92262 Fontenay-aux-Roses, France
7. Laboratoire de micro-irradiation, de métrologie et de dosimétrie des neutrons, PSE-Santé/SDOS, IRSN, 13115 Saint-Paul-Lez-Durance, France
8. CEA, DAM, DIF, F-91297 Arpajon, France
9. Université Paris-Saclay, CEA, LMCE, 91680 Bruyères-le-Châtel, France
10. University of Bordeaux, Centre Lasers Intenses et Applications, CNRS, CEA, UMR 5107, F-33405 Talence, France

We present the first results of proton and X-ray generation from the short-focal-length area (SFA) of the Apollon laser facility, which was performed with the secondary laser beam (F2, 11J, 24 fs), with a nominal power of 0.5 petawatt (PW) [1] and the main laser beam (F1, 45 J, 22 fs), with a nominal power of 2 PW. With a double plasma mirror (DPM) configuration [2], the pre-pulse of F2 can be improved, and thus leads to stronger proton acceleration, with the cutoff energy increases from 25 MeV to 35 MeV. Even though theDPM didn't work well enough for F1, stable acceleration of protons (above 50 MeV) and strong emission of X-rays were recorded, showing good laser-to-target coupling efficiency. These efforts will be followed in2024 by a last commissioning stage at the 10 PW level.

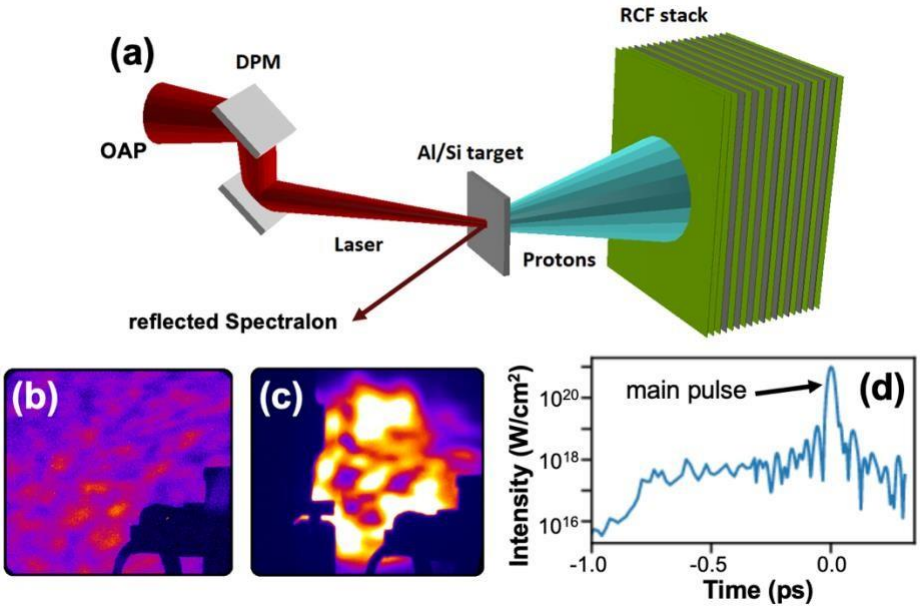


Figure 1 (a) Schematic view of the experimental setup. Images of the specularly reflected laser beam obtained with a Spectralon plate during a direct shot with a 1.5 µm thick Al target (b) and a DPM shot with a 100 nm Si + 50 nm Al target (c). Shadows are present on the images, comingfrom objects placed between the target and the Spectralon plate. (d) Laser contrast measurement during the direct shots (i.e., w/o the DPM).

References:

[1] K. Burdonov, et al., "Characterization and performance of the Apollon short-focal-area facility following its commissioning at 1 PW level." Matter and Radiation at Extremes 6, no. 6 (2021). https://doi.org/10.1063/5.0065138

[2] P. Antici, et al., "Energetic protons generated by ultrahigh contrast laser pulses interacting with ultrathin targets." Physics of Plasmas 14, no. 3 (2007). https://doi.org/10.1063/1.2480610

SESSION 3

High Intensity IR

Jakob Andreasson

ELI-Beamlines

Laplace HC : un accélérateur laser-plasma conçu pour les applications

Alice Vernier

[1] *LOA, Palaiseau*

L'accélération laser-plasma est envisagée depuis deux décennies comme un remplacement potentiel des accélérateurs conventionnels ou comme source d'électrons aux caractéristiques temporelles uniques. Alors que le domaine arrive à maturité scientifique pour l'application à des problématiques de société, des défis majeurs en termes de fiabilité et de stabilité de faisceau restent encore à surmonter.

C'est dans ce contexte que le projet Laplace Haute Cadence a été conçu, avec pour objectif la maîtrise d'un faisceau d'électrons d'une centaine de MeV, une dizaine de pC à 100Hz. Un tel projet suppose d'une part le développement d'un laser aux performances uniques *(100Hz, 300mJ à 1J, 22fs FWHM après compression)* et d'autre part l'élaboration d'un système expérimental optimisé pour la génération de faisceaux d'électrons stables. Cette présentation mettra en perspective les enjeux scientifiques ayant motivé la genèse du projet, notamment l'importance de la haute cadence ainsi que les développements des dernières années au LOA, les chantiers en cours, et les réalisations à venir.

Slowing down the light in vacuum with intense laser pulses

Adrien E. Kraych*[,1], Aurélie Max Mailliet[1], Ali Arras[1], Scott Robertson[1,2], François Couchot[1], and Xavier Sarazin[1]

[1] *Universite Paris-Saclay, CNRS/IN2P3, IJCLab, 91405 Orsay, France*
[2] *Universite Paris-Saclay, Institut d'Optique Graduate School, CNRS, Laboratoire Charles Fabry, 91127 Palaiseau, France*

*krayc h@ijclab.in2p3.fr

Quantum electrodynamics (QED) predicts that vacuum should be a nonlinear optical medium:the speed of light in vacuum should decrease when the vacuum is stressed by intense electro- magnetic fields. This optical phenomenon is similar to the optical Kerr effect in a material medium. Furthermore, this astonishing property of vacuum has never been observed before [1].

The DeLLight (Deflection of Light by Light) experiment aims at measuring this effect using high intense femtosecond laser pulses delivered by the LASERIX platform (E = 2.5 J, 30 fs, 10 Hz) located at IJCLab laboratory (Université Paris-Saclay). The innovative method of DeLLight is to measure the refraction of a laser pulse (probe) of low energy, induced by the index gradient of the vacuum produced by an external pulse (pump) of high intensity. The refraction of the probe pulse is then amplified by a Sagnac interferometer and measured at the output of the interferometer with a CCD camera.

The interferometric measurement technique of the DeLLight experiment was validated dur- ing our last measurement campaign by measuring the deflection of light by light in air using a low-energy pump pulse (μJ).

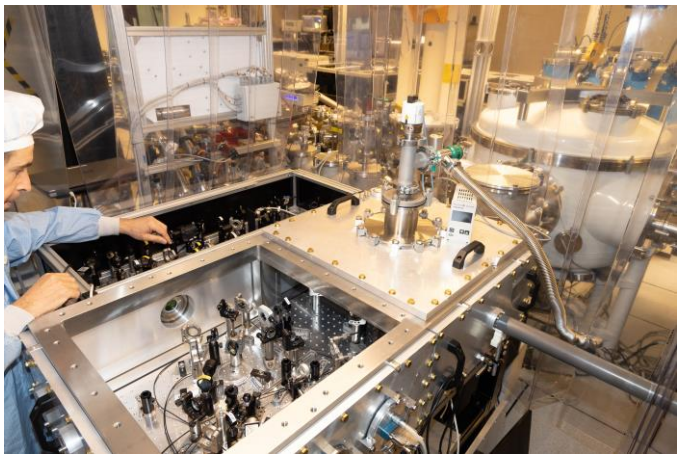


Figure 1: The DeLLight pilot experiment

References:

[1] S. Robertson, A. Mailliet, X. Sarazin, F. Couchot, E. Baynard, J. Demailly, M. Pittman, A.Djannati-Ata¨ı, S. Kazamias and M. Urban (2021). Physical Review A, 103(2), 023524.

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

V. Nefedova, G. Gallé, C. Sire, F. Sylla

SourceLAB SAS, 7 rue de la Croix Martre, 91120 Palaiseau, France

L. Daniault, J. Kaur, R. Lopez-Martens

Laboratoire d'Optique Appliquée, 181 Chemin de la hunière, 91762 Palaiseau, France

E**. Aktan [1,2], C. Greb [1], R. Adam [1], A. Dickson [1,3], B. Hidding [3], M. Büscher [1,2]**

[1]Forschungszentrum Jülich GmbH, PGI 5, Wilhelm Johnen Strasse, 52425 Jülich, Germany

[2]Heinrich-Heine-Universität Düsseldorf, Universitätsstrasse. 1, 40225 Düsseldorf, Germany

[3]Department of Physics, University of Strathclyde, Glasgow G4 0NG, UK

The novel KAIO-Beamline was designed to address the use of laser-plasma accelerators (LPA) for scientific and societal applications. Its modular design incorporates (i) an industrial-grade high average power laser system, (ii) an efficient temporal post-compression stage, based on multi-pass cell technology [1], to reach optimal electron acceleration conditions in the few- cycle [2], and (iii) a compact electron accelerator module with integrated user interface and data management system. The KAIO-Beamline approach is compatible with a wide range of commercial laser platforms.

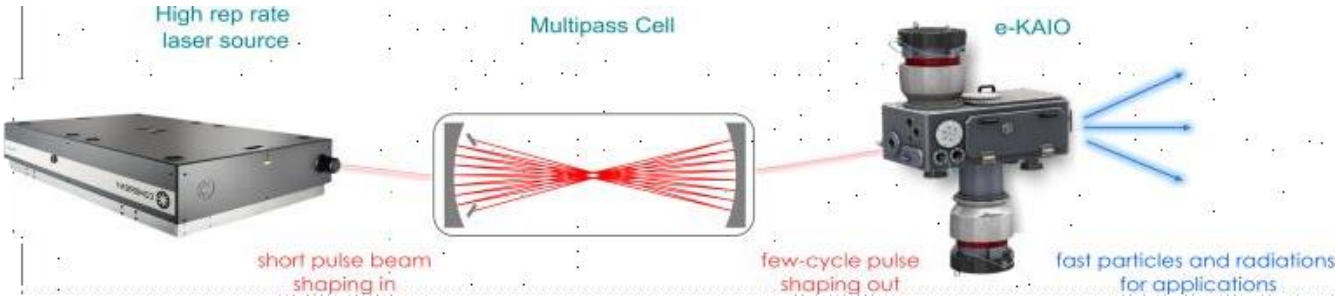


Figure 1: The KAIO-Beamline: the industrial laser system (e.g. ASTRELLA Ti:Sapphire laser from Coherent Inc.) (a) is compressed to produce few-cycle pulses (b) and then sent into the compact table-top e-KAIO source (c) for electron and radiation generation.

Here we will present the first performance results of the KAIO-Beamline using a commercial ASTRELLA Ti:Sapphire laser (Coherent Inc.), delivering 40 fs pulses at 1 kHz repetition rate with energies up to 7 mJ. The laser pulses are comprehensively characterized with novel spatio-temporal metrology tools, such as INSIGHT [3] and TIPTOE [4] techniques.

References

[1] L. Daniault, *et al.*, Opt. Lett. 46, 5264 (2021)
[2] D. Guénot *et al.*, Nature Photonics 11(5), 293-297 (2017)
[3] A. Borot and F. Quéré, Opt. Express **26**, 26444 (2018)
[4] W. Cho *et al.*, Sci Rep **9**, 16067 (2019)

SESSION 4

Condensed phase

Probing the organization of liquids from the bulk to the interface by second harmonic generation

G. Le Breton,[1] O. Bonhomme,[1] A. Pardon,[1] C. Loison,[1] E. Benichou[1]

1. Institut Lumière Matière, UMR CNRS 5579, Université Lyon1, bâtiment A. Kaslter, 43 bv du 11 Novembre 1918, 69622 Villeurbanne Cedex, France

Liquid molecular organization as well in the bulk or at interfaces like gas-liquid or liquid-liquid interfaces, is involved in many physical, chemical, or medical relevant processes. At interfaces, the inherent asymmetry of the forces present dramatically modifies their properties and determines in large part its specific chemical composition, molecular structure, dielectric and transport properties, compared with its bulk counterparts. This asymmetry also drives important chemical reactions occurring in chemical and biological processes, including liquid extraction, catalysis, membrane processes, or drug delivery, to mention a few. In the bulk, this organization is also of utmost importance to understand hydration and more generally solvation, ionsdissolution and precipitation, protein folding and activity, all result from the interaction of ions with solvent molecules. Charged solutes and ions in particular interact with dipolar molecules through ion-dipole interactions and perturb the dipole-dipole interaction network present in pure dipolar liquids. The structure and the dynamics of liquids have been extensively studied with experimental methods based on neutrons, X-rays or UV-to-IR optical spectroscopy as well as theoretical methods like molecular dynamics. Morerecently, nonlinear optical methods [1, 2,3] and Hyper Raman spectroscopy [4] have been proposed withgreat success. The existence of long-range orientational correlations over nanometer distances have been experimentally demonstrated in water as well as the role of ions in disrupting this network [2]. In this context, we have used Second Harmonic Generation (SHG) as a tool to probe the structure of liquids from interfaces to the bulk phase. Indeed, this non-linear optical process whereby two photons at a fundamental frequency are converted into one photon at the harmonic frequency, is intrinsically sensitive to interfacesand very sensitive to the liquid structure or the electrostatic environment.

In this work, we applied the SHG technique through two experimental geometries: surface SHG to studygas-liquid interfaces and Second Harmonic Scattering to study bulk phases. Two kinds of liquids have been investigated: aqueous solutions (pure water and electrolytes) and ionic liquids. We will show that the combination of SHG and quantum mechanical calculations provides valuable information on the structure of the liquids on scales ranging from the very short distance [5] (solvation shells) to the long distance (orientation correlations) [3].

References:

[1] Y. Chen, H.I. Okur, N. Gomopoulos, C. Macias-Romero, P.S. Cremer, P.B. Petersen, G. Tocci, D.M. Wilkins, C. Liang, M. Ceriotti, S. Roke, Electrolytes induce long-range orientational order and free energy changes in the H-bond network of bulk water, Sci. Adv. 2, e1501891 (2016)

[2] J. Duboisset, P.F. Brevet, Salt-induced Long-to-Short Range Orientational Transition in Water, Phys. Rev.Lett. 120, 263001 (2018)

[3] A. Pardon, O. Bonhomme, C. Gaillard, P.F. Brevet, E. Benichou, Nonlinear optical signature ofnanostructural transition in ionic liquids, J. Mol. Liq., 322, 114976 (2021)

[4] M. Okuno, Hyper-Raman spectroscopy of polar liquids excited at 1064 nm: Acetone, acetonitrile, chloroform, and dimethyl sulfoxide, J. Chem. Phys. 152, 174202 (2020)

[5] G. Le Breton, O. Bonhomme, E. Benichou, C. Loison, Liquid Water: When Hyperpolarizability Fluctuations Boost and Reshape the Second Harmonic Scattering Intensities, J. Phys. Chem. Lett., 14, 4158 (2023)

Ultrafast Energy Transfer in Photosynthetic Antenna: Quantumness vs Vibrational Assistance

Manuel LLANSOLA-PORTOLES, CRCN CNRS

Université Paris-Saclay, CEA, CNRS, Institute for Integrative Biology of the Cell (I2BC), 91198Gif-sur-Yvette, France

Photosynthesis is the process which converts the energy of solar photons into an electrochemical potential that, on a slower timescale, is used to reduce CO_2 to sugar. During the first steps, light-harvesting complexes absorb solar photons, and then a cascade of energy transfer leads the resulting excitation energy to a reaction center. In optimal conditions, this process, which involve hundreds of excitation transfer steps, occurs with a quantum yield close to unity. The mechanisms underlying this process and in particular the reasons why the transfers are faster than classical theories predict, are still a matter of very hot debate. Ultrafast time resolved absorption and *2D electronic spectroscopy (2DES)* experiments have shown that the pigment-pigment excitation energy transfers and the first steps of charge separation are modulated by undulations (beats) over more than 2 ps. This was firstly interpreted as beats between coherently coupled states, suggesting that the excitation energy is probing more than a path at once. However, the apparent matching between these beats and the vibrational modes of the cofactors can be interpreted as arising from '*vibrational assistance'*. In this other model, the intersection between the excited state energy surface correspond to the energy of a vibrational mode, and the oscillations observed originate from the movement of the wavepacket in the vibrational excited sub-state of the excited electronic level.

Despite the enormous interest that has arisen in the last decade, it remains unclear whether these beats play a role in energy transfer, and there is a lack of experiments to settle their quantum or vibrational origin. Currently, we are designing and implementing a Femtosecond Stimulated Raman Spectroscopy (FSRS) apparatus with Raman pump tunability. This system will provide the selectivity to probe the vibrational features of each of the excited states evolving in parallel. It will offer detailed information on the excited state structures involved in the photosynthetic process. Subsequently, we will use this information to understand the origin of the beats and model the rates of pigment-to-pigment transfers.

Ultrafast spectroscopy to explore Multiple Exciton Generation

Chloe Magne[1,5], Simona Streckaite[2], Eduardo Domínguez-Ojeda[3], Andrea Echeverri[4], FlavioSiro Brigiano[4], Minh-Huong Ha-Thi[5], Marius Fanckevičius[2], Vidmantas Jašinskas[2], Annamaria Quaranta[1], Andrew A. Pascal[1], Thomas Pino[5], Bruno Robert[1], Julia Contreras-García[4], Daniel Finkelstein-Shapiro[3], Vidmantas Gulbinas[2], Manuel J. Llansola-Portoles[1]

[1]Université Paris-Saclay, CEA, CNRS, Institute for Integrative Biology of the Cell (I2BC), 91198Gif-sur-Yvette, France.

[2]Department of Molecular Compound Physics, Center for Physical Sciences and Technology,Saulėtekio Avenue 3, LT-10257 Vilnius, Lithuania

[3]Insituto de Química, Universidad Nacional Autónoma de México, 04510 Mexico City, Mexico [4]Sorbonne Université, CNRS, Laboratoire de Chimie Théorique, LCT, F. 75005 Paris, France [5]Université Paris-Saclay, CNRS, Institut des Sciences Moléculaires d'Orsay, 91405 Orsay, France.

A promising approach to enhance the efficiency of solar energy collection involves the utilization of Multiple Exciton Generation processes. Among them, Singlet Fission is a mechanism where a photo-excited singlet state divides into two spin-triplet excited states, reaching a theoretical quantum yield of 200%. This is a spin-allowed ultra-fast process thatoccurs in a few picoseconds. One of the main challenges to reaching high quantum efficiencies is the separation of intermediate species, which recombine in picosecond time (e.g., entangled triplets and charge-separated states). In this work, we have investigated singlet fission occurring in water-soluble perylene. We used traditional spectroscopy techniques, including steady-state absorption, fluorescence analysis and ns-to-ms transient absorption. However, for a comprehensive understanding of the system's dynamics, we measured ultra-fast transient absorption (fs-to-ns) and time-resolved fluorescence. To conciliate the data, we applied advanced analysis techniques, such as Target model.

SESSION 5

Materials 1

Non-adiabatic Lifshitz transition in High T_c superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$

J. Dai[1,3], L. Hellbrück[1,2,7], M. Puppin[1,4], A. Crepaldi[1,3], E. Martino[5], L. Yong[6], A. Magrez[6],L. Forró[5], M. Grioni[1,3], F. Carbone[1,2] and S. Benhabib[1,2]

[1] Lausanne Centre for Ultrafast Science (LACUS), École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne

[2]Institute of Physics, Laboratory for Ultrafast Microscopy and Electron Scattering, École Polytechnique Fédérale de Lausanne(EPFL), CH-1015 Lausanne

[3] Institute of Physics, Laboratory of Electron Spectroscopy, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne

[4] Institute of Chemical Science and Engineering, Laboratory of Ultrafast Spectroscopy, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne

[5] Laboratory of Physics of Complex Matter, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne

[6] Plateforme de Cristallogenèse et Caractérisation IPHYS-CGCP, École Polytechnique Fédérale de Lausanne (EPFL), CH- 1015 Lausanne,

[7] Institute of Physics, Laboratory for Quantum Magnetism, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne

Abstract

The equilibrium tunning of doping generates substantial changes in the electronic states of cuprates. They undergo a gradual transition from Mott insulator to Fermi liquid, crossing d- wave superconductivity. Usually, these changes are accompanied by an abrupt transformationin Fermi surface topology, the so-called Lifshitz transition. Here in this work, we address the effect of ultrashort pulses on the Fermi surface topology of cuprates $Bi_2 Sr_2 CaCu_2 O_{8+\delta}$ by means of time-resolved-Angle resolved Photoemission Spectroscopy with pump energy of 1.55 eV. For the first time, we demonstrate that high fluence pulses are significantly efficient in supplying the Fermi level with additional carriers through the photodoping process, driving non-adiabatically the Fermi surface from hole-like to electron-like.

Ultrasmall and tunable TeraHertz surface plasmon cavities at the ultimate plasmonic limit

Ian Aupiais[1], Romain Grasset[1], Tingwen Guo[1], Dmitri Daineka[2], Javier Briatico[3], Sarah Houver[4], Luca Perfetti[1], Jean-Paul Hugonin[5], Jean-Jacques Greffet[5], Yannis Laplace[1]

[1]LSI, CEA/DRF/IRAMIS, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, Palaiseau, France
[2]LPICM, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, Palaiseau, France
[3]Unité Mixte de Physique, CNRS, Thales, Université Paris Saclay, 91767 Palaiseau, France
[4]Université Paris Cité, CNRS, Matériaux et Phénomènes Quantiques, F-75013, Paris, France
[5]Université Paris-Saclay, Institut d'Optique Graduate School, CNRS, Laboratoire Charles Fabry, 91127 Palaiseau, France

The ability to confine THz photons inside deep-subwavelength cavities promises a transformative impact for THz light engineering with metamaterials and for realizing ultrastrong light-matter coupling at the single emitter level. To that end, the most successful approach taken so far has relied on cavity architectures based on metals, for their ability to constrain the spread of electromagnetic fields and tailor geometrically their resonant behavior. In this talk, I will present a different approach allowing for a comparatively high level of confinement by exploiting a plasmonic mechanism based on localized THz surface plasmon modes in bulk semiconductors [2]. We achieve plasmonic confinement at around 1 THz into record breaking small footprint THz cavities exhibiting

mode volumes as low as $V\mathrm{cav}/\lambda 3{\sim}10{-}7{-}10{-}8$, excellent coupling efficiencies and a large frequency tunability with temperature. Notably, we find that plasmonic-based THz cavities can operate until the emergence of electromagnetic nonlocality and Landau damping, which together constitute a fundamental limit to plasmonic

confinement. This work discloses nonlocal plasmonic phenomena at unprecedentedly low frequencies and large spatial scales and opens the door to novel types of ultrastrong light-matter interaction experiments thanks to the plasmonic tunability.

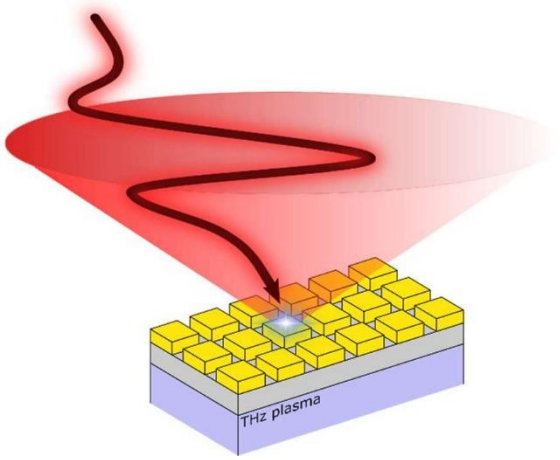


Fig. 1. Sketch of THz photons entering a deeply sub-wavelength THz plasmonic cavity

References

[1] F. Schlawin, D. M. Kennes and M. A. Sentef, Applied Physics Reviews, vol. 9, p. 011312, 2022
[2] I. Aupiais, R. Grasset, T. Guo, D. Daineka, J. Briatico, S. Houver, L. Perfetti, J.-P. Hugonin, J.-J. Greffet, Y. Laplace (to appear in Nature communications)

* Acknowledgement(s): this work was supported by Agence Nationale de la Recherche (grant n° ANR- 17-CE30-0011-11) and Labex PALM (grant n° ANR-10-LABX-0039-PALM).

Single-shot imaging of microscopic turbulent phenomena in 2D quantum fluid-of- light

C. Hainaut[1], C. Ouarkoub[1], S. Ravet[2], J. Bloch[2], P. Suret[1] and A. Amo[1]

[1] Université de Lille, CNRS, UMR 8523 – PhLAM – Physique des Lasers, Atomes et Molécules, Lille, France
[2] Université Paris-Saclay, CNRS, Centre de Nanosciences et de Nanotechnologies, 91120, Palaiseau, France

Out-of-equilibrium quantum gases, especially in two dimension gives rise to complex questions due to the particular den- sity of states and specific connectivity providing thermal and/or quantum fluctuations more importance than in 3-dimensio- nal systems. A prominent example is the low-dimensional quantum turbulent behavior of quantum gases possessing com- plex microscopic dynamics through proliferation of interacting quantized vortices leading to various possibilities of energy redistribution at macroscales.

We investigate an out-of-equilbrium quantum fluid-of-light made of hybrid Exciton-polariton quasiparticles hosted in semi- conductor microcavity [1]. By making use of ultrafast nonlinear imaging technic [2] we are able to measure singleshot amplitude and phase distribution of the fluid-of-light. These picosecond two dimensional snapshots allow us to reveal the presence of quantum vortices, smooking gun of turbulent phenomena. Thus we investigate the conditions requires to entert the turbulent regime of quantum fluid-of-light.

References

[1] I. Carusotto et al., Rev. Mod. Phys. 85, 299 (2013); D. Caputo et al., Nature Materials 17, 145 (2018).
[2] A. Tikan et al., Nat. Phot. 12, 228 (2018) ; P. Suret et al., Nat Comm. 7, 13136 (2016)

Intense and ultrabroadband THz sources at a 200 kHz repetition ratefor time-resolved nonlinear study of low-energy excitations

Niloufar Nilforoushan

[1]Laboratoire de Physique de l'Ecole Normale Supérieure, Université de Paris, Paris, France

[2]Institute of Experimental and Applied Physics, University of Regensburg, Regensburg, Germany

Terahertz (THz) pulses associated with strong electric fields (> few tens of kV/cm) allow the investigation of nonlinear light-matter interactions [1], THz-induced phase transitions [2], and collective excitations of ordered phases [3]. While current sources of intense THz pulses are mostly driven by femtosecond infrared pulses from low repetition rate laser amplifiers (~ 1 kHz), many THz-based spectroscopic techniques also require a high signal-to-noise ratio that implies increasing the repetition rate of the ultrafast laser. In particular, the intermediate range from 100 kHz to 1 MHz, between conventional amplifiers and oscillators, is particularly interesting but has remained elusive for strong electric field ultrabroadband THz generation.

In my talk, I will present the so far missing (ultra-)broadband and intense THz generation by 23 fs optical pulses centered at 1030 nm at a repetition rate of 200 kHz. First, I show mW-level THz emission with a peak electric field in excess of 100 kV/cm driven from optical rectification innonlinear crystals [4]. Next, I demonstrate the generation of strong THz pulses from low- temperature-grown GaAs interdigitated photoconductive antennas [5]. I discuss the underlying THz generation mechanism and I show that the THz spectra lie in the low-frequency range (<3 THz) which is challenging to address using optical rectification.

Our THz source opens a route towards nonlinear time-resolved THz experiments with high signal-to-noise ratios.

References:

[1] H. A. Hafez *et al.*, "Extremely efficient terahertz high-harmonic generation in graphene by hot Dirac fermions", Nature 561, 507 (2018).

[2] P. Salén *et al.*, "Matter manipulation with extreme terahertz light: Progress in the enabling THz technology," Physics Reports, 836–837, 1–74 (2019).

[3] R. Matsunaga *et al.*, "Light-induced collective pseudospin precession resonating with Higgs mode in a superconductor," Science 345, 1145 (2014).

[4] N. Nilforoushan *et al.*, "Ultra-broadband THz pulses with electric field amplitude exceeding 100 kV / cmat a 200 kHz repetition rate," Optics Express 30, 15556 (2022).

[5] N. Nilforoushan *et al.*, "Efficient THz generation from low-temperature-grown GaAs photoconductive antennas driven by Yb-doped fiber amplifier at 200 kHz repetition rate", Applied Physics Letter, accepted (2023)

SESSION 6

Isolated systems 1

Ultra-fast or ultra-slow dynamics through conical intersections? The case of NO_3

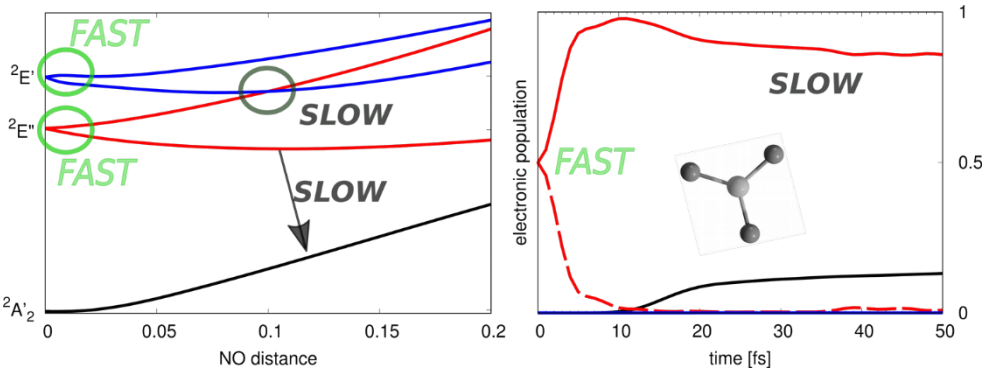
Alexandra Viel[1]

1. Institut de Physique de Rennes, Université de Rennes , UMR CNRS 6251, 35000 Rennes, France

The nuclear dynamics and spectroscopy of the NO_3 radical is still not fully understood despite multiple theoretical and experimental investigations over the last decades. Experimentally, the Neumark group made two breakthroughs by recording in 1991 [1] the first photodetachment spectra of the nitrate anion (NO_3^-) and in 2020 by providing a cryogenic cooling version of these spectra [2]. Their interpretation requires a detailed and complete modelling of the five lowest potential energy surfaces of the radical.

We propose a scheme to simulate photodetachment spectra of NO_3^- from first principles. It relies on the determination of accurate full-dimensional coupled diabatic potential energy surfaces adjusted to high quality ab initio energies via an artificial neural network based scheme [3]. The Muti-Configurational Time Dependent Hartree approach is used to propagate full dimensional wave-packets designed such that temperature effects and the impact of near threshold detachment are taken into account [4].

The two available experiments at high and at cryogenic temperature can be reproduced in very good agreement. A prediction for the photodetachment spectrum in the energy range of the second excited state is proposed [5]. It allows to study the detailed non-adiabatic dynamics by computing the population dynamics. Two behaviors are found: an *ultra-fast* non-statistical radiationless decay amongthe Jahn-Teller components, and a *slow* statistical non-radiative decay among the different state manifolds.



Left: potential energy surfaces along NO distance. Right: electronic populations as a function of time

References:

[1] A. Weaver, et al.; J.Chem.Phys. 94 1740 (1991)
[2] M.C. Babin, et al.; J.Phys.Chem.Lett. 11 395 (2020)
[3] D.M.G. Williams, W. Eisfeld; J.Chem.Phys. 149 204106 (2018)
[4] A. Viel, D.M.G. Williams, W. Eisfeld; J.Chem.Phys. 154 084302 (2021)
[5] D.M.G. Williams, W. Eisfeld, A. Viel; Phys.Chem.Chem.Phys. 24 24706 (2022)

Solvation dynamics of an alkali ion in a helium nanodroplet

Simon H. Albrechtsen[1,6], Constant A. Schouder[2,3,6], Alberto Viñas Muñoz[2], Jeppe K. Christensen[2], Christian EngelbrechtPetersen[1], Marti Pi[4,5], Manuel Barranco[4,5] & Henrik Stapelfeldt[2]

[1] Department of Physics and Astronomy, Aarhus University, Aarhus, Denmark,
[2] Department of Chemistry, Aarhus University, Aarhus, Denmark.
[3] Université Paris-Saclay, CEA, CNRS, LIDYL,Gif-sur-Yvette, France.
[4] Departament FQA, Facultat de Física, Universitat de Barcelona, Barcelona, Spain.
[5] Institute of Nanoscience and Nanotechnology (IN2UB), Universitat de Barcelona, Barcelona, Spain.
[6] These authors contributed equally: Simon H. Albrechtsen, Constant A. Schouder.

Helium nanodroplets are intriguing entities, these superfluid physical objects were made at theend of the 20th century and share common features with Bose-Einstein condensates.
With a temperature of only 0.38K, they are now playing a major role as a host for atoms, molecules and clusters for preparing cold samples and in the formation of atomic/molecular clusters. Their high thermal conductivity, vanishing viscosity and high ionization potential makethem a fantastic solvent for the study of fundamental interactions on a large variety of species.
In this talk, I will show how we can track the successive attachment of helium atoms onto an alkaliion deposited on its surface making it possible to address for the first time a solvation dynamics with an atomistic picture [1]. This approach shifts the usual macroscopic description, through thermodynamics and chemical reaction kinetics, and projects it into its microscopic counterpart, with the attraction and binding of individual atoms to molecules or ions of a solute.

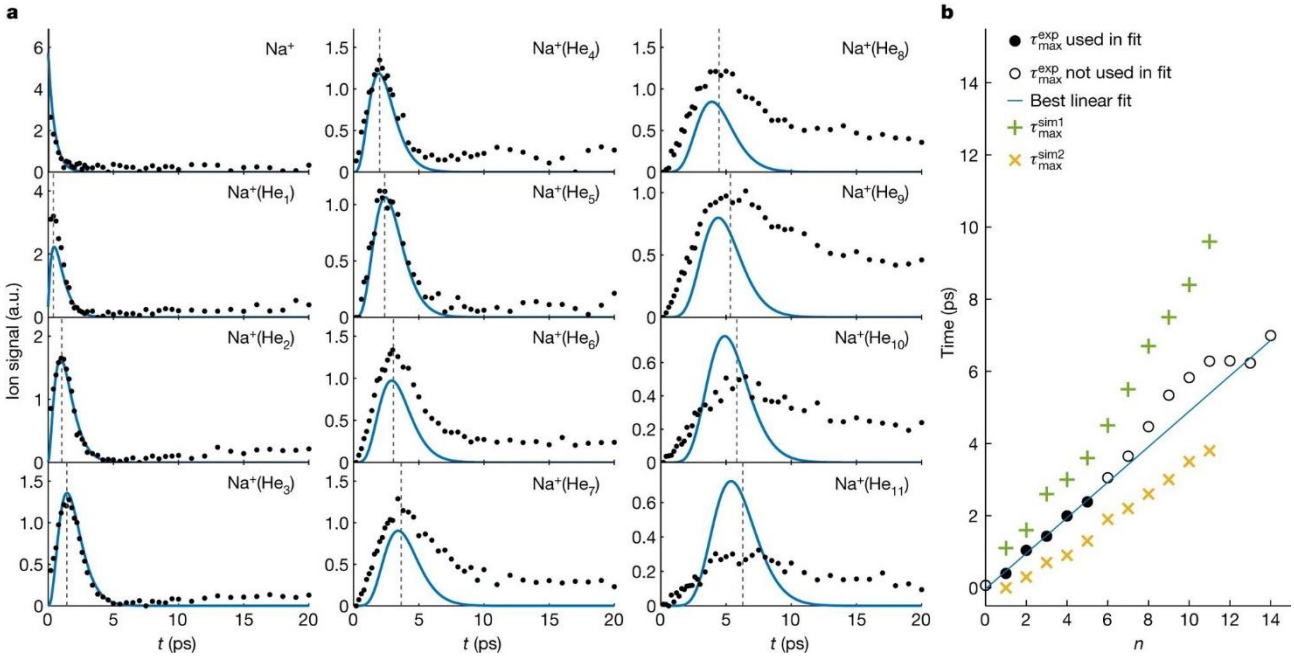


Figure 1: **a**, The black dots represent the time-dependent Na^+He_n ion yield, $Y_n(t)$, for n = 0 to n = 11.The blue curves represent the Poisson probability, $P_n(t)$, that n He atoms have bound to the ion in the interval [0, t], scaled such that the peak of $P_2(t)$ matches the peak of the Na^+He_2 curve. **b**, The time, where each Na^+He_n curve reaches its maximum, marked by the black vertical lines in **a**. The blue line represents the best linear fit to the points with $1 \leq n \leq 5$. The green and orange crosses show the times when n He atoms have moved inside the first or the second solvation shell. Figure taken from [1].

References:

[1] Albrechtsen, S.H., Schouder, C.A., Viñas Muñoz, A. *et al.* Observing the primary steps of ion solvation in heliumdroplets. *Nature* **623**, 319–323 (2023). https://doi.org/10.1038/s41586-023-06593-5

Ultrafast photochemistry of ionized ethylene $C_2H_4^+$: a theoretical perspective

Lina Fransén[1], Thierry Tran[1], Saikat Nandi[2], Morgane Vacher[1]

1. Nantes Université, CNRS, CEISAM UMR 6230, F-44000 Nantes, France
2. Université de Lyon, Université Claude Bernard Lyon 1, CNRS, Institut Lumière Matière, F-69622, Villeurbanne, France

Photoionized and electronically excited ethylene $C_2H_4^+$ can dissociate via H- and H_2-loss, where the latterhas been hypothesized to be preceded by isomerization to the ethylidene form CH_3CH^+.[1] Recent pioneering experiments with few-femtosecond extreme ultraviolet pulses and complementary theoretical studies have shed light on the photodynamics of $C_2H_4^+$.[2-5] However, the details of the mechanisms of H-loss, H_2-loss, and ethylene-ethylidene isomerization, and the factors that govern the competition betweenthese photochemical channels, remain poorly understood.

In this contribution, we simulate the coupled electron-nuclear dynamics of ethylene following ionization and electronic excitation to its four lowest-energy cationic states. The electronic structure is described at the CASSCF level, with an active space large enough to describe bond breaking and formation. This allows us to, for the first time theoretically, reproduce the experimentally observed photochemical pathways (H-loss, H_2-loss, and ethylene-ethylidene isomerization) and shed light on theirrespective mechanisms and time scales.

The electronic relaxation is ultrafast; regardless of the initial electronic state, more than 50% of thepopulation decays within 50 fs to the cationic ground state, where most dissociation and isomerization events occur. Several conical intersections characterized by planar and twisted geometries act as funnels between the low-lying cationic states, and their effects on the photochemical outcome have previously beenhypothesized.[6] We revisit these hypotheses (Figure 1).

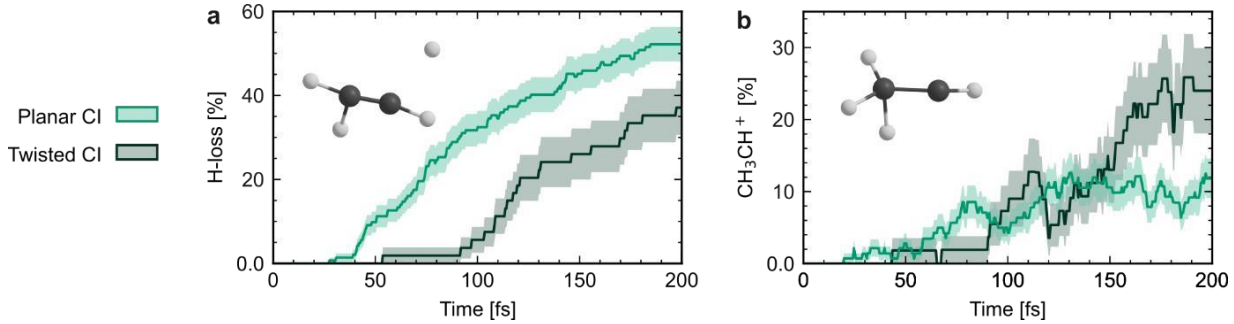


Figure 1. H-loss-yield (a) and ethylidene isomer population probability (b) for the trajectories that transition through planar (light green) andtwisted (dark green) D1/D0 conical intersections.

The simulations moreover suggest that H_2-loss occurs mainly from the ethylidene isomer CH_3CH^+. Thetime scale of ethylene-ethylidene isomerization has previously been inferred in experimental studies employing XUV-pump NIR-probe schemes.[2,3] We find a large discrepancy between the experimentally inferred time scale and that predicted by the simulations and suggest an explanation for this difference.

References:

[1] Sannen, C.; Raşeev, G.; Galloy, C.; Fauville, G.; Lorquet, J. C. *The Journal of Chemical Physics* **1981**, *74*, 2402–2411.
[2] Van Tilborg, J; Allison, T. K.; Wright, T. W.; Hertlein, M. P.; Falcone, R. W.; Liu, Y; Merdji, H; Belkacem, A *Journal of Physics B: Atomic, Molecular and Optical Physics* **2009**, *42*, 081002.
[3] Ludwig, A.; Liberatore, E.; Herrmann, J.; Kasmi, L.; López-Tarifa, P.; Gallmann, L.; Rothlisberger, U.; Keller, U.; Lucchini, M. *The Journal of Physical Chemistry Letters* **2016**, *7*, 1901–1906.
[4] Vacher, M.; Boyer, A.; Loriot, V.; Lépine, F.; Nandi, S. *The Journal of Physical Chemistry A* **2022**, *126*, 5692–5701.
[5] Lucchini, M.; Mignolet, B.; Murari, M.; Gon¸calves, C. E. M.; Lucarelli, G. D.; Frassetto, F.; Poletto, L.; Remacle,F.; Nisoli, M. *The Journal of Physical Chemistry Letters* **2022**, *13*, 11169–11175.
[6] Joalland, B.; Mori, T.; Martínez, T. J.; Suits, A. G. *The Journal of Physical Chemistry Letters* **2014**, *5*, 1467–1471.

Quantum dynamics around ppe's conical intersections for spectroscopic and real-time studies

Joachim Galiana[1], Benjamin Lasorne[1]

1 ICGM, Univ Montpellier, CNRS, ENSCM, Montpellier, France

Keywords: UV-vis spectroscopy, non-adiabatic quantum dynamics, vibronic coupling, conical intersections

In this presentation, we discuss the use of non- adiabatic quantum dynamics for the study of different poly(phenylene)ethynylenes (PPEs), building blocks of light- harvesting dendrimers. PPEs exhibit strongly non-adiabatically coupled electronic excited states. Because of this and of the presence of conical intersections in the potential energy surfaces (PESs) of the molecules, the Born- Oppenheimer approximation breaks down, which requires molecular dynamics to take into account the coupling between the electronic excited states. We propose linear and quadratic vibronic coupling (L/QVC) diabatic model Hamiltonians for reproducing the PESs of the simple PPEs. Next, we use this diabatic potential and inter-state couplings to run quantum dynamics calculations using the Multi Configuration Time-Dependent Hartree (MCTDH) formalism.

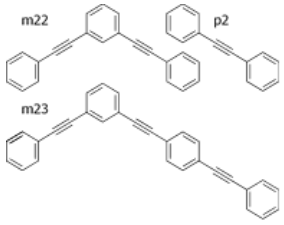


Fig. 1: PPE building blocks of PPE light-harvesting dendrimers

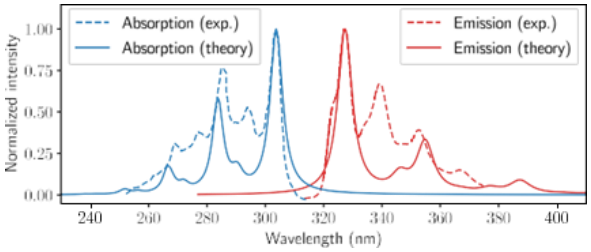


Fig. 2: Stokes-shik contribution to the absorption and emission spectra for the m22 PPE, from experiment[1] and quantum dynamics calculations[4]

The first building block of interest is the symmetrical meta-substituted phenylene (m22, Fig. 1, upper left). The absorption spectra of m22 and its localized fragment (p2, Fig. 1, upper right) are analogous, with similar vibrational structure, which have been measured[1] and reproduced numerically[2,3]. However, their emission spectra are different, with m22 exhibiting an unusual Stokes shift of about 2000 cm^{-1}. In a recent work [4], we identified this Stokes-shifted spectrum as a contribution resulting from the strong inter-state coupling in the first two electronic excited states (Fig. 2). We hereby provide a reasonable explanation for the unusual Stokes shift in the stationary spectroscopy experiments.

The second building block of interest in an asymmetrical meta-substituted PPE (m23, Fig. 1, bottom) and its symmetrical counterpart with two p2 branches, for which excitation-energy transfer (EET) occurs thanks to different branch lengths hence a significant energy gradient. Using analogous Hamiltonian models, we are able to reproduce the expected ultra-fast (< 25 fs) EET from an excitation on the shortest branch toward the longest branch

References:

1. Chu, Q., Pang, Y., Spectrochimica Acta Part A. 2004, 60, 1459-1467.
2. Ho, E. K.-L. ; Etienne, T., Lasorne, B. The Journal of Chemical Physics. 2017, 146, 164303.
3. Ho, E. K.-L. ; Lasorne, B., Computational And Theoretical Chemistry. 2019, 1156, 25-36.
4. Galiana, J. ; Lasorne, B., The Journal of Chemical Physics, 2023, 158, 124113.

SESSION 7

Materials 2

Ultrafast Hidden Spin Polarization Dynamics of Bright and Dark Excitons in 2H-WSe_2

Mauro Fanciulli[1,2], David Bresteau[2], Jérôme Gaudin[3], Shuo Dong[4], Romain Géneaux[2], Thierry Ruchon[2], Olivier Tcherbakoff[2], Ján Minár[5], Olivier Heckmann[1,2], Maria Christine Richter[1,2], Karol Hricovini[1,2] and Samuel Beaulieu[3]

1. Laboratoire de Physique des Matériaux et Surfaces, CY Cergy Paris Université, 95031 Cergy-Pontoise, France
2. Université Paris-Saclay, CEA, CNRS, LIDYL, Gif-sur-Yvette, 91191, France
3. Université de Bordeaux-CNRS-CEA, CELIA, UMR5107, F33405 Talence, France
4. Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China
5. University of West Bohemia, New Technologies Research Centre, 301 00 Plzen, Czech Republic

We performed spin-, time- and angle-resolved photoemission spectroscopy (STARPES) [1] with extreme ultraviolet (XUV, 35.65 eV) from the high harmonic generation beamline FAB10 of Attolab [2] on the inversion-symmetric bulk 2H-WSe2 transition metal dichalcogenide (TMDC). Excitons were prepared in the system by photoexcitation with infrared (800 nm) circularly polarized light [3]. The very short probing depth of XUV photoemission permits selective measurement of photoelectrons originating from the top- most WSe2 layer, allowing for direct measurement of hidden spin polarization [4,5] of bright and momentum-forbidden dark excitons [3]. Our results [6] reveal efficient chiroptical control of bright excitons' hidden spin polarization. Following optical photoexcitation, intervalley scattering between nonequivalent K-K' valleys leads to a decay of bright excitons' hidden spin polarization. Conversely, the ultrafast formation of momentum-forbidden dark excitons acts as a local spin polarization reservoir, whichcould be used for spin injection in van der Waals heterostructures involving multilayer TMDCs.

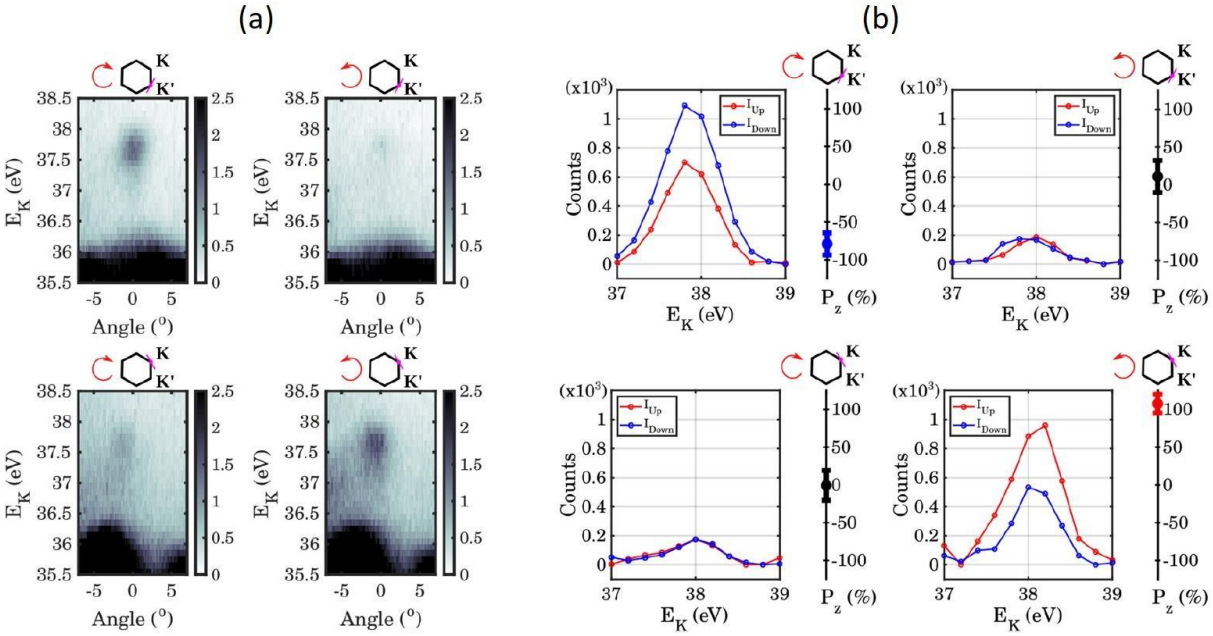


Fig.1. STARPES measurements on 2H-WSe2. (a) Bandmaps at pump-probe overlap showing the formation of bright excitons, for different pumppulse helicities (circular right or left red arrow) and electron valleys (K and K' in the hexagonal Brillouin Zone), as depicted on top of each panel. Magenta lines indicate the analyzer's slit direction. (b) Spin-resolved energy distribution curves (EDC) for the four cases of (a), showing in red andblue the up and down spin channels along the sample's normal direction. The amount of spin polarization is indicated on the right of each panel.

References:

[1] M. Fanciulli et al., Phys. Rev. Res. 2, 013261 (2020)
[2] D. Bresteau et al., Eur. Phys. J. Spec. Top. 1 (2023)
[3] S. Dong et al., Natural Sci. 1, e10010 (2021)
[4] X. Zhang et al., Nat. Phys. 10, 387 (2014)
[5] E. Razzoli et al. Phys. Rev. Lett. 118, 086402 (2017)
[6] M. Fanciulli et al., Phys. Rev. Lett. 131, 066402 (2023)

Out-of-equilibrium dynamics in the Bi2212 antiferromagnetic phase

L. Gatuingt[1], A. Alekhin[1], M. Cazayous[1], A. Sacuto[1], G. Gu[2], S. Houver[1], Y. Gallais[1]

1. Université Paris-Cité, Laboratoire Matériaux et Phénomènes Quantiques, CNRS (UMR 7162), 75013 Paris, France
2. Matter Physics and Materials Science, Brookhaven National Laboratory, Upton, New York 11973, USA

Ultrafast time-resolved Raman spectroscopy is applied to study the antiferromagnetic (AF) phase in the cuprate high-Tc superconductor Bi2212. The focus is on the evolution of Raman active magnetic and electronic excitations after an ultrafast 60fs near-infrared pump pulse. We observe a significant sub- picosecond decrease of the AF two-magnon excitation intensity, proving that the AF phase is altered by the pump pulse. Using Raman selection rules, we discuss the separate effects of the pump pulse on the AF order and on electronic carriers. Finally, we compare these effects with that of hole doping.

References:

[1] Kota Katsumi, Alexandr Alekhin, Sofia-Michaela Souliou, Michael Merz, Amir-Abbas Haghighirad, Matthieu Le Tacon, Sarah Houver, Maximilien Cazayous, Alain Sacuto, and Yann Gallais, Phys. Rev. Lett. 130 . 106904, 2023.

[2] Yao Wang, Thomas P. Devereaux, and Cheng-Chien Chen Phys. Rev. B 98, 245106, 2018

Electron-phonon coupling and ultrafast dynamics of hot carriers in semiconductors: from interpretation of photoemission experiments totransport simulations in devices

Jelena Sjakste, Raja Sen, Nathalie Vast

Laboratoire des Solides Irradiés, CEA-DRF-IRAMIS, Ecole Polytechnique, CNRS UMR 7642, Institut Polytechnique de Paris, 9112O Palaiseau, France

Electron-phonon coupling determines the charge transport properties of materials as well as the relaxation dynamics of photoexcited carriers. Computational methods based on density functional theory, on the onehand, and time-energy- and momentum- resolved spectroscopy, on the other hand, allow today an unprecedently detailed insight into the role of the electron-phonon coupling [1]. At the same time, hot carriers start to attract attention in the context of emerging concepts for energy conversion.

In this work, we will present the theoretical and experimental results for relaxation dynamics of photoexcited electrons in semiconductors [2-5]. Our computational method based on density functional theory (DFT) and on interpolation of the electron-phonon matrix elements in Wannier space allowed to successfully interpret the dynamics of photoexcited electron relaxation in GaAs, Si, Ge and InSe, in good agreement with two-photon photoemission experiments [2-5]. In particular, recently we have studied, by photoemission spectroscopy and by ab initio calculations, the influence of the two-dimensional (2D) electron gas, created on the InSe surface by Cs deposition, on the relaxation dynamics of photoexcited electrons. We have shown that the remote coupling with the bulk phonons persists even in the case of complete thermalization of the photoexcited electrons with the 2D gas, and that the coupling of electrons to polar optical phonons is not completely screened even at high Cs concentrations [5].

More generally, we will discuss the necessity of time-resolved simulations as well as the problematics relatedto the coupling of *ab initio* data with device-oriented simulation methods. Indeed, the search for new functional thermoelectric and/or photovoltaic materials requires device-oriented simulation methods of carrier transport, such as stochastic Monte Carlo method, as well as *ad hoc* information on the strength andrelative importance of the various scattering channels, which can only be obtained from *ab initio* calculations[6].

References:

[1] J. Sjakste, K. Tanimura, G. Barbarino, L. Perfetti, and N. Vast, J. Phys: Cond. Mat. **30**, 353001 (2018).
[2] H. Tanimura, J. Kanasaki, K. Tanimura, J. Sjakste, N. Vast, M. Calandra, F. Mauri, Phys. Rev. B 93, 161203(R) (2016).
[3] Z. Chen, C. Giorgetti, J. Sjakste, et al. Phys. Rev. B 97, 241201(R) (2018).
[4] H. Tanimura, J. Kanasaki, K. Tanimura, J. Sjakste, and N. Vast, Phys. Rev. B **100**, 035201 (2019).[5] Z. Chen, J. Sjakste et al, PNAS **117**, 21962-21967 (2020).
[6] R. Sen, N. Vast, J. Sjakste, Appl. Phys. Lett. **120**, 082101 (2022).

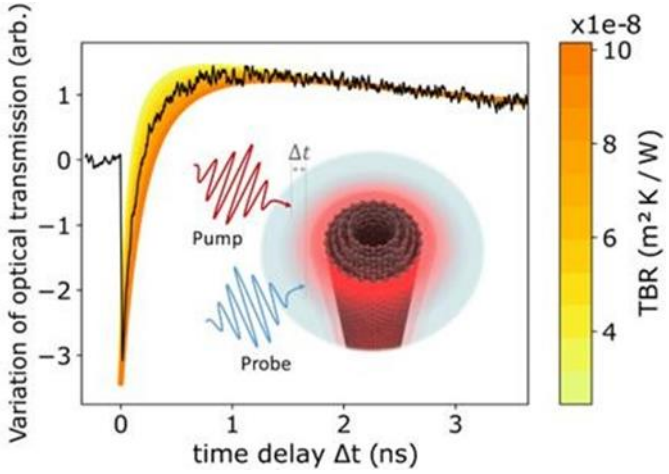
Experimental Measurement of Nanoscale Heat Transfer with Ultrafast Optical Spectroscopy

Alessandro Casto, Margherita Vittucci, Fabien Vialla, Aurelien Crut, Natalia Del Fatti, Francesco Banfi and Paolo Maioli

P. Maioli is with the FemtoNanoOptics group at Institut Lumière Matière (iLM), Université de Lyon, CNRS, Université Claude Bernard Lyon 1, 69622 Villeurbanne, France
A. Casto, M. Vittucci, M. Diego, F. Vialla, A. Crut, F. Vallée, N. Del Fatti, F. Banfi are with the FemtoNanoOptics group at Institut Lumière Matière (iLM), Université de Lyon, CNRS, Université Claude Bernard Lyon 1, 69622 Villeurbanne, France.
F. M. Bellussi and M. Fasano are with Politecnico di Torino, Department of Energy, 10129 Torino, Italy

paolo.maioli@univ-lyon1.fr

We use ultrafast optical transient absorption spectroscopy to measure the thermal dynamics of metal nanoparticles and carbon nanotubes in solid and liquid environments, thus allowing quantitative determination of the Thermal Boundary Resistance at the interface. In this technique, a laser pulse (pump) induces ultrafast thermal excitation of the nano-objects. Their cooling dynamics is optically monitored using a second time-delayed optical pulse (probe). This work opens the way to fundamental investigations of nanoscale heat transfer and applications to thermal management in nanocomposite materials.



Experimental transient absorption signal on multi-wall CNTs in water (black) and its fits with a complete opto-thermal model (colors). Different colors correspond to different values of Thermal Boundary Resistance. In the inset, a pictorial representation of the time-delayed pump and probe technique.

*Research supported by Institut Universitaire de France (IUF).

SESSION 8

Secondary sources 1

PI-FROSt: "Plasma-Induced Frequency Resolved Optical Switching"

E. Hertz[1], R. Bhalavi[1,2], A. Leblanc[3], F. Billard[1], O. Faucher[1], P. Béjot[1]

1. Laboratoire Interdisciplinaire Carnot de Bourgogne, UMR CNRS 6303 Université de Bourgogne 21078 DIJON CEDEX - France
2. Femto Easy, Batiment Gienah, Cité de la Photonique, 11 avenue de Canteranne, 33600 Pessac France
3. Laboratoire d'Optique Appliquée, Ecole Polytechnique, ENSTA, CNRS, Université Paris Saclay, Palaiseau, France

A new characterization technique for ultrashort laser pulse metrology providing a phase-matching free pulse retrieval is presented. The method, called PI-FROSt for "Plasma-Induced Frequency resolved Optical Switching" is based on the laser-induced plasma defocusing phenomenon. In PI-FROSt, a plasma lens is induced by focusing a pump pulse in a rare gas (here Ar). A probe pulse propagating through the plasma will then be defocused, increasing its size in the far field. This phenomenon is used as a switch by inserting a coronagraph in the probe path that lets some part of the probe passing around only when the medium is ionized. The signal propagating around the coronagraph is then redirected to a spectrometer that records the signal spectrum as a function of the pump-probe delay. The resulting spectrogram allows a complete characterization of the temporal (and spectral) characteristics of the probe pulse as with the standard FROSt method [1]. The potentiality of PI-FROSt is evaluated through the characterization of fs laser pulses in the near-infrared (l=805 nm) and UV (l=266 nm) spectral domains with a pump pulse centered at l =805 nm. Typical results are depicted in Figure 1.

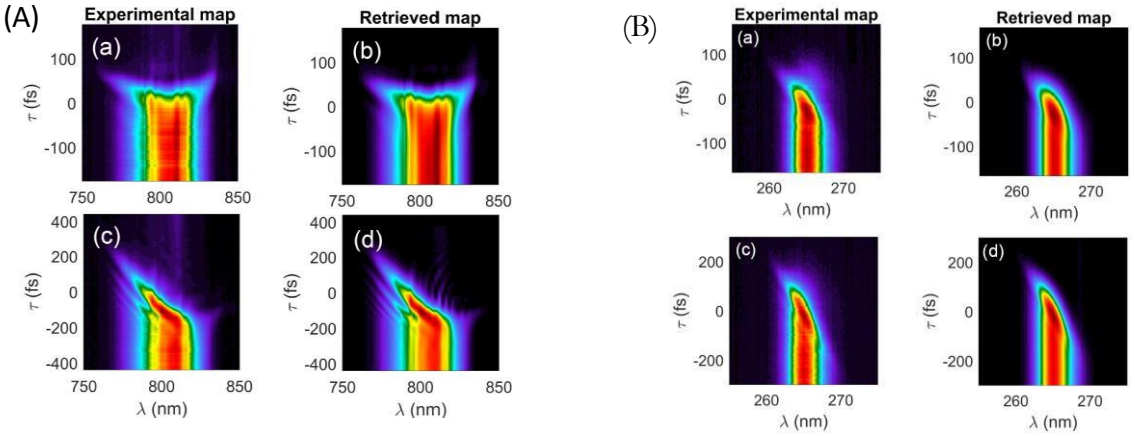


FIGURE 1

PI-FROSt measurements. (a) Experimental and (b) reconstructed spectrograms for a compressed probe pulse. (c, d) same as (a, b) for a probe pulse after propagating through a dispersive plate. Left part (A) corresponds to a probe pulse in the NIR spectral region and right (B) for a probe pulse in the UV spectral region.

The assessment of the method demonstrates laser pulse reconstructions of high reliability in both near- infrared and ultraviolet spectral ranges. The method is simple, free of phase matching constraints and can potentially operate over an exceptionally broad spectral range, ultimately limited by the gas absorption in the UV range (≈ 120 nm for the first excited state of Ar) and by the plasma frequency in the far-IR range (≈ 100 μm). PI-FROSt features several other advantages since it has no damage threshold and can inherently operate with pump and probe beams of same wavelength in both self- or cross-referenced configurations. In addition, the relaxation of the PI-FROSt signal has been measured to approximately 600 ps in Ar anticipating applicability at ultra-high repetition rates, possibly reaching GHz frequencies (while offering the possibility of measuring pulse durations up to several tens of picoseconds). Although additional tests conducted under more challenging conditions are needed to assess possible limitations of the method, the aforementioned strengths of PI-FROSt demonstrate its potential as a valuable tool for characterizing ultrafast sources with non-standard wavelengths or bandwidths.

References:

[1] A. Leblanc et al., Opt. Express 27, 20 (2019)

Simple few-shot method for spectrally resolving the wavefront ofan ultrashort laser pulse

S. Smartsev[1,3], A. Liberman[2,3], I. A. Andriyash[1], A. Cavagna[1], A. Flacco[1], C. Giaccaglia[1], J. Kaur[1], J. Monzac[1], S. Tata[2], A. Vernier[1], V. Malka[2], R. Lopez-Martens[1], J. Faure[1]

1. Laboratoire d'Optique Appliquée, ENSTA Paris, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, 828 Bd. des Maréchaux, 91120 Palaiseau, France
2. Department of Physics of Complex Systems, Weizmann Institute of Science, 234 Herzl St., Rehovot 7610001, Israel
3. The authors contributed equally to this work

We present a novel and experimentally simple method for measuring the multispectral wavefronts of ultrashort laser pulses in either a single shot or a few shots. IMPALA, or Iterative Multispectral Phase Analysis for LAsers, relies on a customized iterative algorithm and a specialized pinhole mask [1]. The arrangement of pinholes is optimized to ensure that the multi-color interference patterns created by any pair of holes do not overlap with others in the spatial Fourier plane. This configuration enables the retrieval of spectrally resolved wavefronts of the ultrashort beam, spatially sampled at the pinhole positions in a single shot. Additionally, rotations of the mask allow for improved spatial resolution of the wavefronts. Weconducted a proof-of-principle experiment using a 30 fs laser (the spectrum is depicted in Fig. 1 (i)), as shown in Fig. 1. We successfully retrieved a proper amount of pulse front tilt (PFT) introduced in the controlled way through adjustments of the parallelism of the compressor gratings in their dispersion plane(Fig. 1 (ii)). The measured wavefronts for a grating tilt of 2.3' (minutes of arc) and their Zernikedecomposition are presented in Fig. 1.

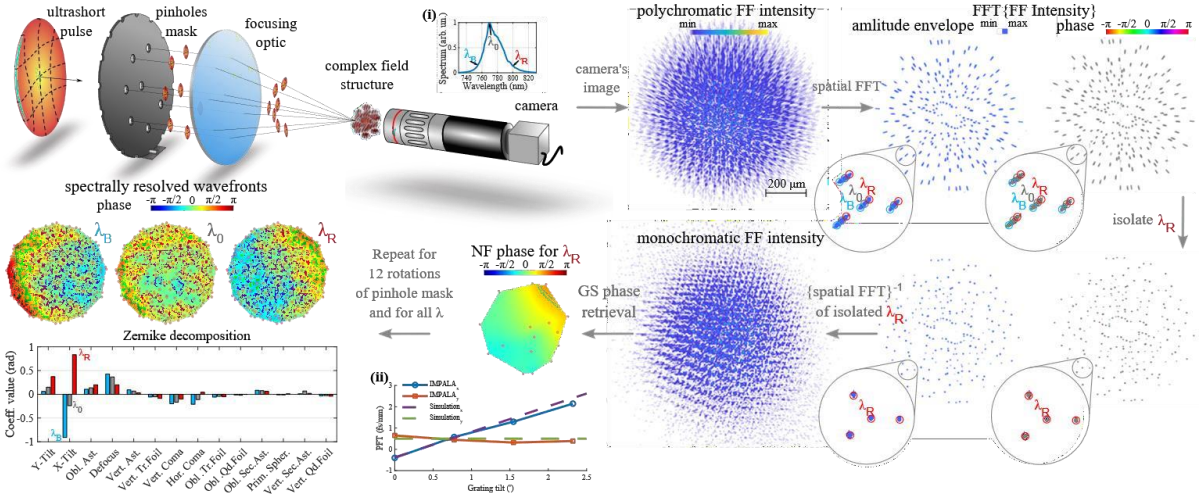


FIG. 1. Simplified experimental setup and wavefront retrieval sequence: A mask splits an ultrashort laser pulse with STCs into dozens of beamlets. The focusing optic concentrates the beamlets at its focal plane, which is then imaged by a microscope objective onto the CCD camera, thus registering the polychromatic FF intensity. The monochromatic FF intensity is extracted from the measured polychromatic FF intensity by utilizing the spatial separation of colors in the spatial FFT plane. A phase retrieval algorithm reconstructs the NF phase of the beamlets at the hole's position using the monochromatic FF intensity. The process is repeated for different colors and mask rotations to obtain spectrally resolved wavefronts, from which the Zernike coefficients of the analyzed wavefronts are extracted.

References:

[1] S. Smartsev et. al, "Simple few-shot method for spectrally resolving the wavefront of an ultrashort laser pulse," arXiv:2307.15799, Jul 2023.

Développements récents dans la modélisation pour l'accélération laser-plasma d'électrons

F. Massimo[1], I. Moulanier[1], L. T. Dickson[1], G. Maynard[1], B. Cros[1]

[1] Laboratoire de Physique des Gaz et des Plasmas, CNRS, Université Paris Saclay

Au cours des dernières années, le rôle de la modélisation numérique dans l'accompagnement des expériences d'accélération par sillage laser-plasma d'électrons a progressé de façon significative. Une sélection de résultats récents de modélisation, obtenus par la communauté française du domaine,sera présentée.
Parmi ces résultats on montrera en particulier le rôle crucial de la description précise du laser dans lessimulations pour obtenir des accords sans précédent avec les expériences. On présentera les modèles rapides et précis qui ont été mis au point et utilisés pour la conception d'expériences basée sur l'exploitation des données et les perspectives sur l'utilisation de l'apprentissage automatique et de l'intelligence artificielle.
Les perspectives pour aborder les nombreux défis, représentant des enjeux majeurs pour les prochaines années vers la réalisation d'un accélérateur laser-plasma d'électrons multi-étages serontégalement évoquées.

SESSION 9

Isolated systems 2

Ensemble density functional theory of electrons and nuclei

Emmanuel Fromager

Laboratoire de Chimie Quantique, Institut de Chimie de Strasbourg (UMR7177),Université de Strasbourg, Strasbourg, France.

Density-functional theory (DFT) has become over the decades the method of choice for modeling, within the Born-Oppenheimer (BO) approximation, the ground-state electronic structure of molecules (and solids) [1]. This can be explained by the drastic simplification of the electronic structure problem that DFT offers by mapping the exact electronic ground-state density onto a singleelectronic configuration (which is described by the sole occupied molecular orbitals in that configuration). DFT can be extended, in principle exactly, to the beyond-BO simulation of molecules *via* the exact factorization of the molecular wavefunction [2,3]. From a practical point of view, the alternative ensemble DFT approach [4,5,6], which allows for a simultaneous treatment of ground and excited electronic states, has attracted in recent years an increasing attention, in particular for the simulation of non-adiabatic nuclear dynamics [7]. In this talk I will discuss the formal exactification (and the subsequent practical approximations) of such a strategy where nuclear wavefunctions are combined with a many-electron density-functional ensemble, starting from the Born-Huang expansion of the molecular wavefunction. The challenges to be addressed in the time- dependent regime will also be discussed.

References

[1] "DFT Exchange: Sharing Perspectives on the Workhorse of Quantum Chemistry and Materials Science"- Andrew M. Teale, TrygveHelgaker, Andreas Savin, et al., Phys. Chem. Chem. Phys. (2022) 24, 28700- 28781.
[2] Villaseco Arribas, E.; Agostini, F.; Maitra, N.T., Molecules 2022, 27, 4002.
[3] Requist, R.; Gross, E.K.U., Phys. Rev. Lett. (2016) 117, 193001.

[4] E.K. Gross, L.N. Oliveira, and W. Kohn, Phys. Rev. A 37,
2809 (1988).[5] E. Fromager, Phys. Rev. Lett. 124, 243001 (2020).

[6] F. Cernatic, B. Senjean, V. Robert, and E. Fromager, Top Curr Chem (Z) 380, 4 (2022).
[7] M. Filatov, S. K. Min, and K. S. Kim, J. Chem. Theory Comput. 2018, 14, 4499-4512.

Probing two-photon resonant ionization with electroninterferometry

L. Maëder[1], R. Menezes Ferreira[1], HJB. Marroux[1], C. Schouder[1], M. Lejman[1], A. Autuori[1], D. Platzer[1], N. Berrah[2], R. Taïeb[3], J. Caillat[3], P.Salières[1]

[1] *Université Paris-Saclay, CEA, CNRS, LIDYL, 91191, Gif-sur-Yvette, France*

[2] *Department of Physics, University of Connecticut, Storrs, Connecticut, 06269, USA*

[3] *Sorbonne Université, CNRS, Laboratoire de Chimie Physique-Matière et Rayonnement, France*

rafael.menezes-ferreira@cea.fr

XUV coherent sources based on high-order harmonic generation are a powerful tool to access temporal information on ionization processes through two-photon interferometric schemes. In particular, the RABBIT technique is widely used to reconstruct ionization dynamics with both attosecond time resolution [1] and spatial resolution [2].

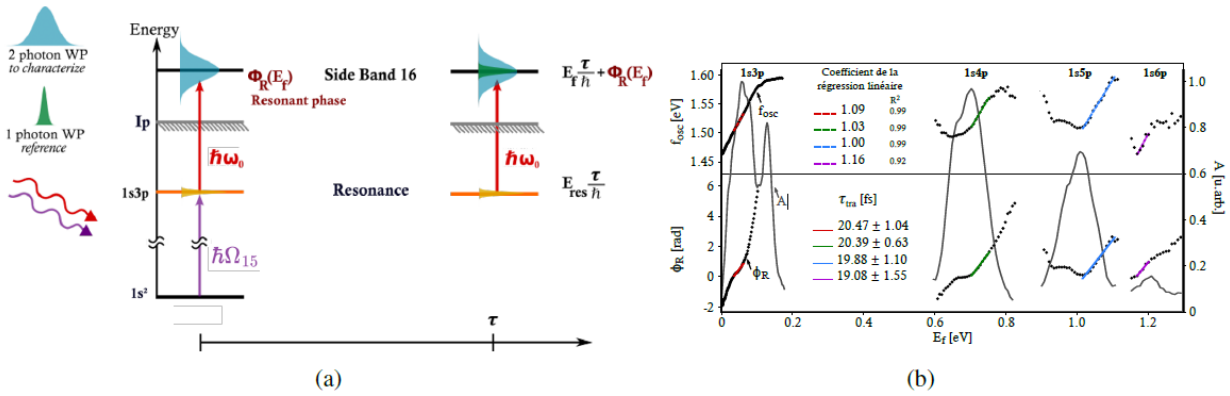


Figure 1: a) Interferometric scheme of the experiment; b) Retrieved phases in multiple 1snp resonances of Helium

Here, we present the temporal study of two-photon resonant ionization of Helium using an original interferometric scheme, alternative to RABBIT. Helium atoms are ionized by simultaneous absorption of harmonic15 (H15) and the fundamental IR, creating the electron wavepacket (EWP) of interest. When H15 is close to resonance with a 1snp state, the latter is coherently populated. At a later time, after a delay τ, a second IR pulse ionizes the excited helium by absorption of an IR photon, providing a reference EWP that interferes withthe EWP of interest. Their spectral interference encodes their relative phase that contains the phase Φ_R of theresonant two-photon transition (the spectral derivative of which gives the transition delay):

$$\Delta\varphi(E_f,\tau) = \frac{E_f - E_{1s3p}}{\hbar}\tau - \Phi_R(E_f) \qquad (1)$$

The experiments evidence a transition delay of 20 fs due to the transient trapping of the electron on the resonance before ionization. As compared to the RABBIT technique [2], this scheme offers a high contrast of the spectral fringes and a very precise temporal information. Indeed, since the reference EWP results from two sequential one-photon transitions, it does not introduce any phase/delay and thus can be considered as 'ideal'.

References

[1] M. Isinger *et al.*, Photoionization in the time and frequency domain, *Science* 356, 898-896 (2017)

[2] A. Autuori *et al.*, Anisotropic dynamics of two-photon ionization: An attosecond movie of photoemission, *Science advances* 8, eabl7594 (2022)

Quantum interference effects in the post-ionization dynamics of fluoro-benzene unravelledthrough non-adiabatic simulations

Anthony Ferté and Morgane Vacher

Laboratoire CEISAM, Nantes Université, CNRS, Nantes, France

Controlling matter with light has always been an exciting idea that led to the development of the ever-expanding field of photochemistry. Moreover, with the emergence of techniques to generate ultrashort attosecond light pulses, whose importance has just been rewarded by the 2023 NobelPrize in physics, hope was born to go beyond the possibilities of standard photochemistry. Indeed, due to their large energy bandwidth, such pulses can populate coherent electronic wavepackets. Theprospect of exploiting the interference between components of such a wavepacket to steer chemical reactivity is the core concept of attochemistry.

As chemical reactions and photo-induced processes are dynamical in essence, accurately simulatingthe evolution in time of molecular systems is often the key to fully revealing the characteristics of the process at play. Vertical inoization of the fluoro-benzene molecule leads very close to the conical intersection between the two lowest doublet cationic states (see figure 1.a) makig this systeme a formidable choice to investigate quantum coupled electron-nuclear dynamics induced upon ionization by attosecond/sub-femtosecond pulses.

We will see that, as shown in figure 1.b, by tuning the composition of the cationic wavepacket populated upon ionization of the fluoro-benzene molecule, one can fully control the subsequent dynamics of the system [1]. I will especially focus on the identification of quantum interferences that drive the dynamics and I will show that they leave clear signatures in the shape of the autocorrelation function (figure 1.c), a property that can be accessed experimentally using high harmonic spectroscopy.

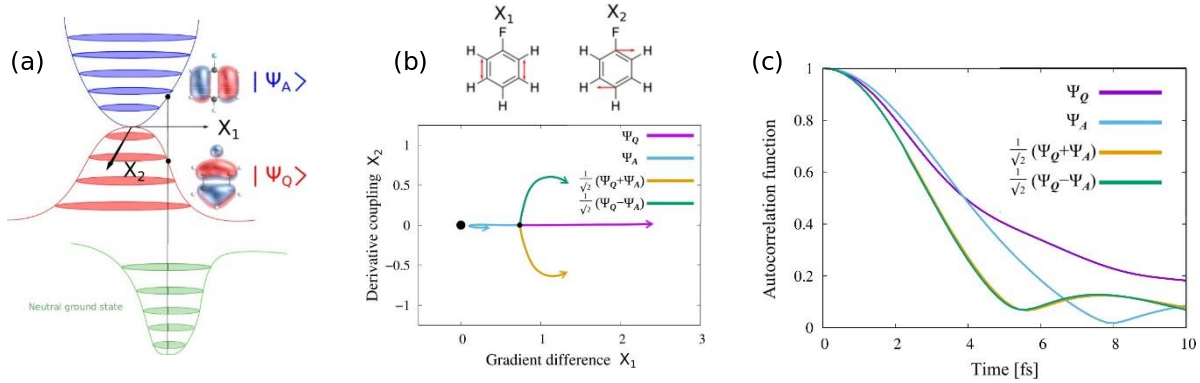


Figure 1: (a) Schematic representation of the fluoro-benzene vertical ionization. (b) Average quantum nuclear motion and (c) autocorrelation functions upon excitation of different initial cationic wavepackets [1].

[1] A. Ferté, J. P. Malhado, and M. Vacher, arXiv, 2309.08269

SESSION 10

Isolated systems 3

Theoretical study of electron-nuclei entanglementin molecular photoionization

Morgan Berkane, Camille Lévêque , Richard Taïeb , Jérémie Caillat

Sorbonne Université, CNRS, Laboratoire de Chimie Physique-Matière et Rayonnement, LCPMR, Paris, France

Attosecond time-resolved experiments allow the observation « in real time » of photoemission[1], driven by the correlated interactions between the photoelectron and its parent ion. They hence provide unprecedented insight on this process at its most fundamental level. These experiments are essentially based on phase measurements by means of interferometric schemes. In their common implementations, they assume coherent processes, in particular at the level of the pump/probe light sources but also of the studied phenomena themselves. Here we study signatures of decoherence induced by incomplete measurements in time resolved molecular photoionisation using numerical simulations on a simple model molecule.

We are interested here in molecular photoionization where the correlations between the photo-electron and the vibrational dynamics of the ion are likely to create entanglement [2]: the totalsystem can then no longer be written as the product of the two subsystems, which translates into a loss of coherence in partially resolved measurements.

Using simulations on diatomic model molecules composed of an active electron and the vibrational degree freedom, we investigate how decoherence manifests in XUV molecular photoemission, inspired by a previous theoretical work [2]. We focus on how this loss of coherence translates into experimental observables and ultimately into their interpretation in terms of ultrafast dynamics. More precisely, we consider the case of molecular RABBIT [3] and mixed-FROG [4] and compare the actual dynamics revealed in time-dependent numericalsimulations to the dynamics reconstructed from electron spectra or ion yields « measured » atvarious levels of resolution.

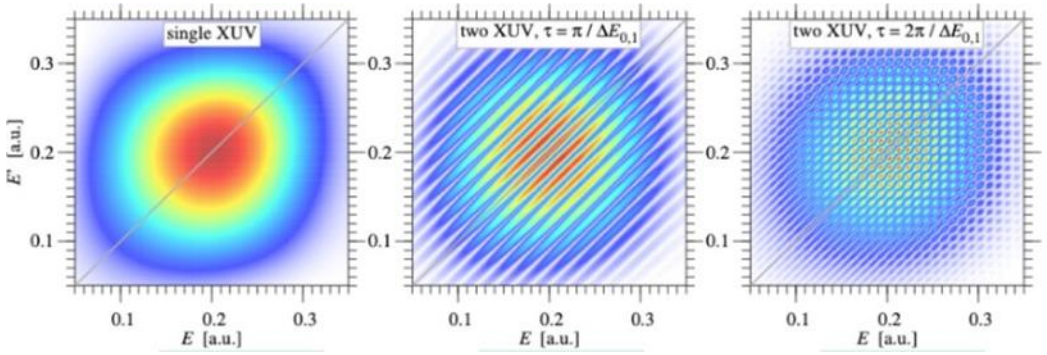


Figure 1 : Photoelectron reduced density matrix obtained when ionizing our model moleculewith a pair of 21.7 eV pulses with various delays (see also [2]).

References:

[1] Desrier A, Maquet A, Taïeb R, and Caillat J Phys. Rev. A 98, 053406 (2018)

[2] Vrakking Marc J. J. Phys. Rev. Lett. 126, 1 13203(2021)

[3] Caillat J, Maquet, A, Haessler S, Fabre B, Ruchon T, Salières P, Mairesse Y, and Taïeb R Phys. Rev. Lett. 106, 093002 (2011)

[4] Bourassin-Bouchet C, Barreau L, Gruson V, Hergott JF, Quéré F, Salières P, and Ruchon T Phys. Rev. X 10, 031048 (2020)

Radiationless phenomena with in the exact factorization

Francesco Talotta

Université de Lorraine, Laboratoire de Physique et Chimie Théoriques UMR7019, 57070, Metz, France

Radiationaless transitions are important non-adiabatic phenomena that take place in excited molecular systems. Depending on the type of electronic states involved and the type of state-to-state couplings, these transitions can be classified into internal conversions (ICs) and intersystem crossings (ISCs). The former involves states of the same spin multiplicity interacting via the non-adiabatic couplings (NACs),whereas the latter involve states of different spin multiplicity interacting via the relativistic spin-orbit couplings (SOCs).
A proper description of these non-radiative transitions is of utmost importance in the study of the photophysical and photochemical properties of molecular systems, as these two phenomena are notoriously responsible for ultrafast relaxations processes. Therefore, a number of computational methods have been developed in the last decades to simulate non-adiabatic dynamics with trajectory- based algorithms, such as the very popular trajectory surface hopping (SH) [1], or the novel coupled trajectory mixed quantum/classical (CT-MQC) method [2].
In my presentation, I will first briefly review the main ideas behind SH and CT-MQC, pointing out the main differences and current applicability of both methods. Then, I will focus on the CT-MQCalgorithm, on the latest development and extension of this method with the inclusion of the relativistic spin-orbit interactions, to let CT-MQC deal with both NACs and SOCs on the same footing [3,4]. A brief review of the theory and implementation of SOCs in CT-MQC will be presented, together with a detailed assessment of the new algorithm, through application on model systems that can be easily compared with the exact quantum wavepacket dynamics. The results of both CT-MQC and exact calculations will be critically discussed, underling the very good agreement between the two methods, but also the limitations of the current implementation. A detailed explanation of the reasons behind the limitations will be also presented.

[1] M. Barbatti, *WIREs Comput. Mol. Sci*, **2011**, 1, 4, 620.
[2] F. Agostini et al., *J. Chem. Theory Comput.* **2016**, 12, 5, 2127.
[3] F. Talotta et al., *Phys. Rev. Lett.*. **2020**,124, 033001.
[4] F. Talotta et al. *J. Chem. Theory Comput.* **2020**, 16, 4833.

Towards table-top and high-flux OPCPA drivers at 2.1 µm for Soft-X-Ray generation.

Raman Maksimenka[1], Nicolas Thiré[1], Thomas Pinoteau[1], Simon Pomarède[1], Nicolas Forget[2], Yoann Pertot[1]

1 Fastlite by Amplitude, 165 rue des Cistes, Antibes, France
2 Universite Cote d'Azur, CNRS, Institut de Physique de Nice (INPHYNI),
yoann.pertot@amplitude-laser.com

By combining industrial-grade solid-state lasers at 1030 nm with nonlinear conversion processes, one can build powerful driver sources in the infrared with favorable properties for driving HHG in in the soft-X-Ray region. Optical parametric chirped-pulse amplifiers (OPCPAs) are ideal systems for producing the desired parameters for such experiments: a combination of long wavelength, few-cycle pulse duration, carrier-envelope phase (CEP) stability, high peak intensity, and high average power.

Here, we present OPCPA architectures based on supercontinuum seeding, delivering sub- 25fs pulse durations, sub-100 mrad rms non-averaged CEP stabilities, tens of W of average power and mJ-level energies with excellent spatial properties. Design considerations as well as characterization methods to achieve such record performances are presented in detail. Furthermore, power and energy scaling based on state-of-the art Ytterbium pump lasers are also discussed.

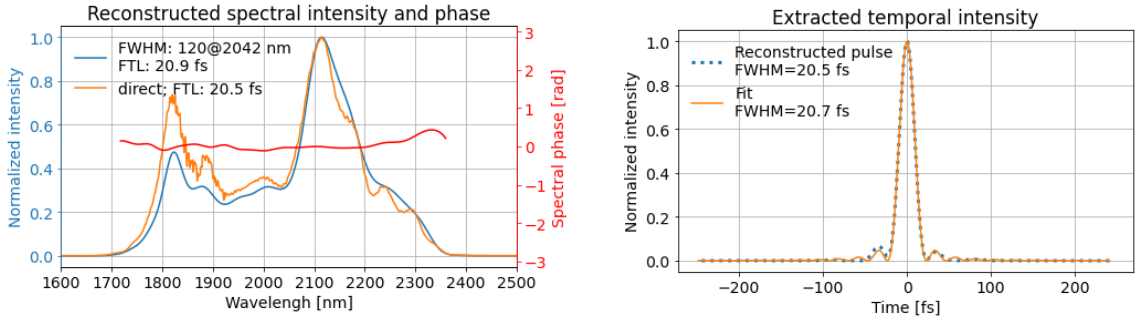


Figure 1 Pulse duration measurement at 2.1 µm, by self-referenced spectral interferometry. Top panel: directly measured spectral intensity (orange line, left-hand Y scale) and reconstructed spectral phase (red line, right-hand Y scale) and spectrum (blue line; left-hand Y scale; Bottom panel: extracted temporal intensity.

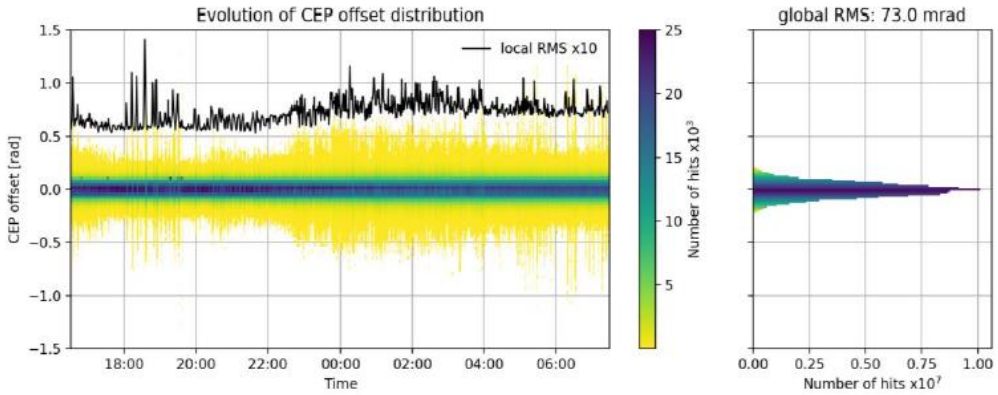


Figure 2: 16 hours-long non-averaged CEP measurement at 2.1 µm. global RMS: 73 mrad

SESSION 11

Secondary sources 2

Circumventing limits of Laser-Plasma Acceleration with advanced optics

Igor Andriyash

LOA, Palaiseau

For the last decades, the development of Laser-Plasma Accelerators (LPAs) has attracted high interest thanks to the capacity of plasma to produce and sustain extremely high electric fields. The accelerating gradients in plasma accelerators can exceed 100 GV/m, which is three orders of magnitude larger than those obtained in metallic-cavity accelerators, thus promising very compact alternatives to conventional linear machines [1]. However, a high field is not the only ingredient required for high multi-GeV energy gains, as the accelerated beam has also to follow this field over long distances. Today the identified main challenges for LPA are the diffraction and depletion of the driver laser, and the dephasing of the accelerated beam with the driven plasma waves. Diffraction and pump depletion cause laser intensity to fall during the acceleration, eventually suppressing the wakefield, while dephasing results from the mismatch between the phase velocity of the accelerating field and the one of the electron beam, and it leads the electron beam towards a decelerating phase of the wake.

Here we discuss two approaches for tackling these limitations and increasing the beam energy. We will present the first experimental demonstration of acceleration of quasi-monoenergetic electron beams at the GeV level in a plasma waveguide created by a quasi-Bessel machining beam shaped by an axiparabola mirror [2,3]. Another concept employs an advanced optical shaping of the laser driver that allows a diffraction-free propagation over a long distance while controlling the group velocity of the laser [4], thus greatly extending the effective dephasing length [5].

References

[1] E. Esarey, C. Schroeder and W. Leemans, Review Modern Physics 81, 1229-1285, (2009)

[2] S. Smartsev, et al. Optics Letter 44, 3414-3417 (2019).

[3] K. Oubrerie, et al. Light Science & Applications 11, 180 (2022)

[4] K. Oubrerie, I. A. Andriyash, R. Lahaye, S. Smartsev, V. Malka and C. Thaury, Journal of Optic 4, 045503 (2022)

[5] C. Caizergues, S. Smartsev, V. Malk

Modelling electron deflectometry measurements of magnetic fields inultrahigh-intensity, femtosecond laser-foil interactions

S. Passalidis[1,2], S. Corde[3], X. Davoine[1,2], M. Gilljohann[3], O. Kononenko[3], Y. Mankovska[3], P. SanMiguel Claveria[3], L. Gremillet[1,2]

[1] CEA, DAM, DIF, 91297 Arpajon, France

[2] Université Paris-Saclay, CEA, LMCE, 91680 Bruyères-le-Châtel, France

[3] LOA, ENSTA Paris, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, 91762 Palaiseau, France

We examine numerically the processes of magnetic field generation in relativistic femtosecond laser-solid interactions. Our study is motivated by a recent experiment at LOA, whereby the B fields induced in a thin (~20 μm) solid foil by a $\sim 10^{19}$ Wcm^{-2}, ~30 fs laser pulse were diagnosed via electron deflectometry. In contrast to a previous experiment[1], the ~100 MeV-range probe beam was produced by an auxiliary laser-wakefield accelerator, and injected into the solid foil through its rear (non-irradiated) surface. The mean angular deflection and root-mean-square (rms) spread of the beam electrons after exiting the irradiated foil surface showed nontrivial dependencies on delay time and transverse position with respect to the driving laser pulse.

We compare these measurements with the results of 2D collisional particle-in-cell simulations run under conditions as close as possible to the actual ones. Notably, we take into account the 2D preplasma created by the laser's pedestal and describe self-consistently the interaction of the probe electrons with the induced plasma fields. Two main B-field generation mechanisms are found to account for the observed electron deflections: (i) the collisionless current filamentation instability[2], which excites strong (>10^3 T), kinetic-scale fields around the laser spot[3]; (ii) the fountain-like motion of the fast electrons near the plasma-vacuum boundaries, which leads to azimuthal B fields surrounding the laser spot up to ~100 μm radii[4,5].

Our synthetic deflectometry maps reproduce qualitatively the experimental data as regards both the mean and rms deflections. To shed further light on the simulation results, we proceed with a quasistatic approach which enables the respective effects of the small- and large-scale field components to be isolated as a function of the location and time of probing.

References

[1] G. Raj et al., Phys. Rev. Res. 2, 023123 (2020).

[2] A. Bret, L. Gremillet, and M. Dieckmann, Phys. Plasmas 17, 12050 (2010).[3] J. C. Adam et al., Phys. Rev. Lett. 97, 205006 (2006).

[4] G. Sarri et al., Phys. Rev. Lett. 109, 205002 (2012).

[5] W. Schumaker et al., Phys. Rev. Lett. 110, 015003 (2013).

Probing strong-field Quantum Electrodynamics with Doppler-boosted laser beams

Luca Fedeli, Pierre Bartoli, Neil Zaïm, Thomas Clark, Henri Vincenti

CEA-Saclay, France

Physical scenarios characterized by electromagnetic fields so strong that quantum electro- dynamics (SF-QED) plays a substantial role are one of the frontiers of contemporary plasma physics research. At LIDYL we study strategies to use optical devices called "plasma mirrors" curved by radiation pressure to boost the intensities of existing ultra-intense lasers by the Doppler effect and focus them to extreme field intensities, high-enough to use them to study SF-QED. In this contribution we present such strategies as well as the numerical tools required to model these physical scenarios. In particular, we will present WarpX, a state-of-the-art, open- source Particle-In-Cell code conceived to address the challenges of computing at the exascale, as well as PICSAR-QED, a portable Monte Carlo module providing WarpX with the capability of simulating the SF-QED phenomena that are usually the most relevant.

POSTERS

Attosecond Raytracing (ART) – A Free Python Code for Simulating Attosecond Beam Transport

André Kalouguine[1], Anthony Guillaume[1], Charles Bourassin-Bouchet[2], and Stefan Haessler[1,*]

[1]Laboratoire d'Optique Appliquée (LOA), Institut Polytechnique de Paris, CNRS, 91120 Palaiseau, France
[2]Laboratoire Charles Fabry, Institut d'Optique Graduate School, CNRS, Univ. Paris-Saclay, 91120 Palaiseau, France

Synopsis We present the open-source python code *Attosecond Raytracing (ART)*, which performs ray tracing simulations with a specific focus on ultrashort (attosecond) laser pulses. The code currently focuses on reflective optics, freely arrangeable including grazing incidence configurations. It can serve for designing optimized optics for the challening refocusing of high-NA attosecond beams, stemming from small source spots and/or targeting high on-target intensities.

In experiments, attosecond light pulses or phase-controlled IR/visible light waveforms are generally used in-focus. The spatio-temporalfocus results from the interference of all spatio- spectral components, governed by their optical path lengths. The extreme temporal precision required in the field of attosecond science dic- tates a corresponding extreme sensitivity to such optical-path differences: 30 nm correspond to a 100-as temporal shift. The resulting spatio- temporal coupling, i.e. a spatial dependence of the temporal profile of a light pulse, can dra-matically increase the focal-averaged duration the intensity-envelope of (attosecond) XUV pulses [1], or blur the focal-averaged phase of IR/visible optical cycles. Together with the as- sociated increase of the focal spot size, the in- focus light intensity drops accordingly.

The reflective beam transport and refocusing optics typically employed in attosecond physics setups (mostly spherical and toroidal mirrors) are aberrating, i.e. they induce optical path length differences. The higher the numerical apertures (NA), or equivalently the divergence angles, of the involved beam sections, the stronger the aberrations. We find that for diver- gence full-angles ≥ 20 mrad, i.e. NA ≥ 0.01, aberrations must be carefully considered.

Diffraction makes such values inevitable whenever the (virtual) source and/or the image-focus is very small. This is the case, e.g. when the driving laser is tightly focused, such as for HHG in gases or solids with low-pulse energy (and high rep-rate) drivers [3,4], or for surface- HHG on plasma mirrors at ultra-high driving intensity [5]. It is also the case in demagnifying setups that boost the attosecond pulse intensity [6-8]. Focusing high-NA attosecond pulses is thus a major technical challenge, first addressed ten years ago in raytracing calculations [1,6].

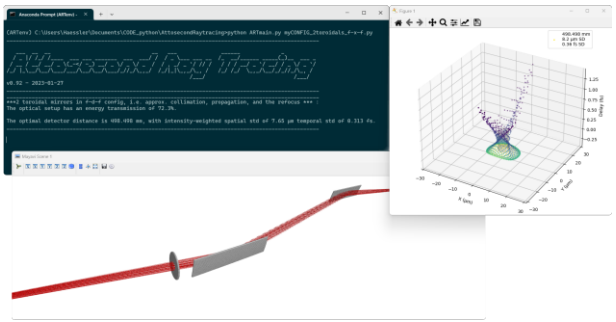


Figure 1. A simuation with ART of a 1:1 imaging via two toroidal mirrors for a 30-mrad beam.

Attosecond Raytracing (ART) now makes such calculations avaible to the ultrafast com-munity in an accessible open-source python code [10]. It lets users freely arrange and orient several reflective optics with common surface shapes (spherical, parabolic, toroidal, ellipti-cal, …). Random surface shape errors with giv- en RMS-value can be added, and the alignment sensitivity of the optical setups determined.

We will demonstrate ART with simulations of existing micro-focusing setups [6-8], as wellas possible re-focusing setups for the Δθ=70- mrad HHG-beams from a relativistic plasma mirrors.

References

[1] Bourassin-Bouchet C *et al* 2010 *JOSA A* **27** 1395**;** 2013 *Opt. Express* **21** 2506
[3] Krebs M *et al* 2013 *Nature Phot.* **7** 555
[4] Gonzalez A I *et al* 2018 *JOSA B* **35** A6
[5] Haessler S *et al* 2022 *Ultrafast Science* **2022** 9893418
[6] Poletto L et al 2013 *Opt. Express* **21** 13040
[7] Coudert-Alteirac H *et al* 2017 *Appl. Sci.* **7** 1159
[8] Nayak A *et al* 2018 *Phys. Rev. A* **98** 023426
[9] Haessler S *et al* 2023 https://mightymightys.github.io/AttosecondRaytracing

2

A New Time- and Polarization-Resolved Momentum Microscopy Apparatus at CELIA

Sotirios Fragkos[1], Samuel Beaulieu[1]

1. Université de Bordeaux - CNRS - CEA, CELIA, UMR5107, F33405 Talence, France

We report on the development of a new beamline and endstation for time- and polarization-resolved momentum microscopy, specifically designed for the investigation of ultrafast out-of-equilibrium processes in quantum materials [1]. Our beamline features ultrafast (~100 fs), high-repetition-rate (250 kHz) monochromatic extreme ultraviolet (21.6 eV) photons with full (linear, elliptical, and circular) polarization tunability. Driving high harmonic generation (HHG) using an annular beam allows us to spatially separate the high-power 515 nm driving laser from the XUV beamlet while preserving the linear polarization axis angle tunability. This enables control of the polarization state of the XUV radiation with a fixed all-reflective phase shifter. The XUV spot size on target is 30 µm x 40 µm FWHM. This beamline is coupled to a time-of-flight Momentum Microscope (GST mbH and Surface Concepts GmbH), allowing for simultaneous detection of the full surface Brillouin zone over an extended binding energy range without the need to rearrange the sample geometry. Spatial resolution of ~1 µm is achievable by inserting an aperture in the Gaussian plane of the microscope. Our instrument also features a retarding electrostatic front lens allowing us to direct secondaries and pump- induced slow electrons back to the sample surface, opening up a previously inaccessible regime of pump fluences in photoemission experiments [2]. This instrument will be used for multi-modal dichroism in XUV photoemission spectroscopy of quantum materials undergoing ultrafast light- induced dynamics.

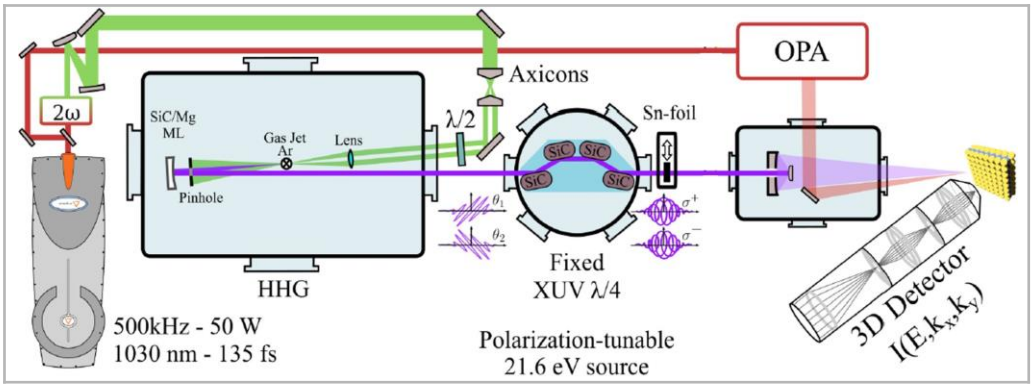


Figure 1: Schematic illustration of the experimental setup developed at CELIA.

References:

(1) A. Comby et al., Ultrafast polarization-tunable monochromatic extreme ultraviolet source at high-repetition-rate, J. Opt. 24, 084003 (2022).

(2) G. Schönhense et al., Suppression of the vacuum space-charge effect in fs-photoemission by a retarding electrostatic front lens, Rev. Sci. Instrum. 92, 053703 (2021).

3

Towards time-resolved nano-crystallography with

Maryam Alashoor,[1] Philippe Rabiller,[1] Maciej Lorenc,[1] Hiroko Tokoro,[2] Shin-ichi Ohkoshi,[3] Florian Banhart, [4] Matthieu Picher, [4] Yaowei Hu[4]

[1] *Institut de Physique de Rennes, UMR CNRS 6251 University of Rennes 1, F-35000 Rennes, France (*email: maryam.alashoor@etudiant.univ-rennes.fr)*

[2] *Department of Chemistry, School of Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan (*email:ohkoshi@chem.s.u-tokyo.ac.jp)*

[3] *Department of Materials Science, Faculty of Pure and Applied Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8577, Japan (*email:tokoro@ims.tsukuba.ac.jp)*

[4] *Institut de physique et chimie des Matériaux de Strasbourg, UMR CNRS 7504 University of Strasbourg, F-67000 Strasbourg, France (*email:Florian.Banhart@ipcms.unistra.fr)*

This work focuses on structural phase transition induced by a laser pulse in tritatnium pentaoxide, by using transmission electron microscopy (TEM) to obtain ultrafast electron diffraction on ferroelastic nano-crystallites of Ti_3O_5. The bulk crystal of Ti_3O_5 is known to exhibit a semiconductor-to-metal phase transition at high temperature, around 450 K, associated with a structural symmetry breaking. However, when a bulk crystal is downsized to the nanoscale (25-500 nm), a metallic phase with the same symmetry as the semiconductor is stabilized, revealing a very broad temperature range of bi-stability, up and beyond the room temperature. This nanoscopic effect arises from the contribution of the surface or interface energy to Gibbs free energy [1]. The nano-crystallites of Ti_3O_5 were synthesized by S.Ohkoshi and H.Tokoro at the School of Science, University of Tokyo [2]. Previous pump-probe structural experiments on this system were carried out at large-scale facilities, and even though atomic resolutions were achieved, the macroscopic insights were limited by averaging over a huge number of crystallites [3]. It is remedied in this study by using bursts of electrons focused onto a single crystallite, as a probe [4]. Reversible switching between the bistable phases (trapped metallic λ-phase and the semiconductor β-phase) can be induced upon light irradiation. Indexation has been made after developing a graphical interface for electron diffraction patterns. However, clear-cut distinction between these phases is still a challenge due to the similarity of lattice parameters. A possible explanation is that multiple crystallites contribute to electron beam scattering, which compromises the diffraction overall resolution in reciprocal space. The next iteration of this experiment is underway, whereby suitably thin crystals of much-improved surface and homogeneity will be put under laser and electron beams.

References

(1) Ohkoshi, Shin-ichi, Yoshihide Tsunobuchi, Tomoyuki Matsuda, Kazuhito Hashimoto, Asuka Namai, Fumiyoshi Hakoe, and Hiroko Tokoro. "Synthesis of a metal oxide with a room-temperature photoreversible phase transition." Nature chemistry 2, no. 7 (2010): 539-545.

(2) Nasu, T., H. Tokoro, K. Tanaka, F. Hakoe, A. Namai, and S. Ohkoshi. "Sol–gel synthesis of nanosized λ-Ti3O5 crystals." In IOP Conference Series: Materials Science and Engineering, vol. 54, no. 1, p. 012008. IOP Publishing, 2014.

(3) Mariette, Céline, Maciej Lorenc, Hervé Cailleau, Eric Collet, Laurent Guérin, Alix Volte, Elzbieta Trzop et al. "Strain wave pathway to semiconductor-to-metal transition revealed by time-resolved X-ray powder diffraction." Nature communications 12, no. 1 (2021): 1239.

(4) Hu, Yaowei. "Ultrafast electron microscopy of transient states in nanomaterials." PhD diss., Université de Strasbourg, 2022.

4

Ultrafast dynamics of a spin-polarized electron gas

Benjamin Bakri[1], Giovanni Manfredi[1], Paul-Antoine Hervieux[1], Nicolas Crouseilles[2],and Xue Hong[2]

[1] *Universite´de Strasbourg, CNRS, Institut de Physique et Chimie des Mate´riaux de Strasbourg, UMR 7504, F-67000 Strasbourg, France*

[2] *Universite´de Rennes, Inria Rennes (Mingus team) and IRMAR UMR CNRS 6625, F-35042 Rennes, France*

Ultrafast magnetism is an actively explored research area in the general framework of ultrafast phenomena. For instance, the demagnetization of a thin magnetic layer by femtosecond laser pulses [1] and its fundamental origin are still under debate. Ultrafast magnetic phenomena are also crucial to devise and implement effective THz emitters.

In order to describe the coupled charge/spin dynamics in a ferromagnet, a great variety of models are available, going from the time-dependent spin density functional theory to quantum hydrodynamic models. An alternative approach consists in modelling the electrons with a phase- space approach similar to that used in plasma physics.

In this approach, the electrons are described by a probability distribution function in the phase-space (x, v) [2]. For particles with spin, four distribution functions are required: one for the density of electrons in the phase space $f0(x, v, t)$, and three for the spin density $fi(x, v, t)$, where $i = (x, y, z)$ denotes the spin direction. These distribution functions obey an evolution equation similar to the Vlasov equation of plasma physics, coupled to a Poisson equation for the mean-field Coulomb potential [3, 4]. This constitute the itinerant part of the magnetism.

In addition, the fixed-ion magnetism is described by the Landau-Lifschitz equation, coupled to the above spin-Vlasov equation through an exchange term of the Rundermann-Kittel-Kasuya- Yosida (RKKY) type. The overall model [5] is schematized in Fig. 1.

Our work is aimed at studying the dynamics of magnons in an infinite ferromagnetic material. In particular, we investigate the effect of the electron dynamics on the damping and propagation of the magnons. We observe that the magnon amplitude is modulated nonlinearly under strong excitation of the electron dynamics.

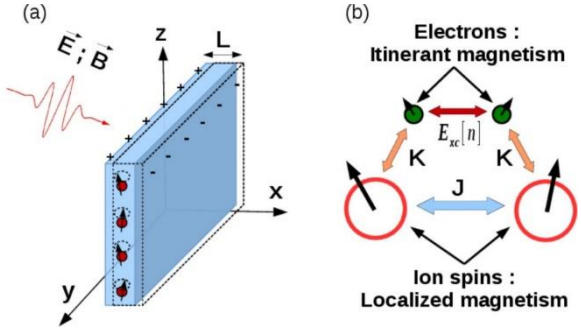


Figure 1: Schematic view of the model coupling the itinerant electron magnetism to the fixed electronmagnetism (right panel), following excitation by an ultrashort laser pulse (right panel). From [5].

References

[1] Eric Beaurepaire, J-C Merle, A Daunois, and J-Y Bigot. Ultrafast spin dynamics in ferromagneticnickel. *Physical review letters*, 76(22):4250, 1996.

[2] Giovanni Manfredi. How to model quantum plasmas. *arXiv: quant-ph/0505004*, 2005.

[3] Giovanni Manfredi, Paul-Antoine Hervieux, and Jérôme Hurst. Phase-space modeling of solid-state plasmas. *Reviews of Modern Plasma Physics*, 3(1):1–55, 2019.

[4] Nicolas Crouseilles, Paul-Antoine Hervieux, Xue Hong, and Giovanni Manfredi. Vlasov–maxwellequations with spin effects. *Journal of Plasma Physics*, 89(2):905890215, 2023.

[5] J Hurst. *Ultrafast spin dynamics in ferromagnetic thin films*. PhD thesis, University of Strasbourg,2017.

5

Ultrafast light-induced strain in the prototypal multiferroic material BiFeO3

V. Juvé[a], R. Gu[a], H. Bouyanfif[b], G. Vaudel[a], A. Poirier[a], M. Weber[a], B. Dkhil[c], C Laulhé[d], Ph Hollander[d] C. Paillard[c], D. Sando[e], S. Fusil[f], V. Garcia[f], P. Ruello[a]

[a]Institut des Molécules et Matériaux du Mans, UMR 6283 CNRS, Le Mans Université, 72085 Le Mans, France,
[b]Laboratoire de la Matière Condensée , UR UPJV 2081, Univ Jules Vernes Picardie, France,
[c]Laboratoire Structures, Propriétés et Modélisation des Solides, UMR CNRS 8580, CentraleSupelec, Université Paris Saclay, Gif-Sur-Yvette, France.
[d]Synchrotron SOLEIL, Université Paris Saclay, 91190 Saint-Aubin, France.
[e]School of Materials Science and Engineering, University of New South Wales, Sydney 2052, Australia,
[f]Unité Mixte de Physique CNRS, Thales, Université Paris-Saclay, Palaiseau 91767, France

*pascal.ruello@univ-lemans.fr

Ultrafast light excitation of a material at a time shorter than any thermalization time between charges, phonons and spins can open new avenue to control the matter and to drive functional materials in new states not achievable in standard thermodynamic equilibrium. The light-induced transient states in func- tionnal materials (magnetic, multiferroics, topological materials, …) remain complex due to multiple cross couplings between these degrees of freedom and the description of the different pathways to es- tablish a quasi-equilibrium for the charges, phonons and spins is nowadays a challenge. Furthermore, driving these materials with light and in the sub THz-THz range is crucial for future fast data processing [1]. To do so, it is necessary to understand and measure the ultrafast dynamics of each of these elemen- tary particles and quasi-particles : charges, phonons and spins. It is well known that ultrafast light pulses lead to a modification of the lattice dynamics at short time scale due to the existence of different electron- phonon and spin-phonon coupling [1-3]. In solids, phonons spectra usually extend over several THz indicating that the coupling between hot electron and out-equilibrium spin can take place rapidly (below 1 ps) after an ultrafast light excitation [1-4]. The role of phonons (coherent, incoherent) has gained more and more interest in femtomagnetism and in multiferroic materials in general [4-6] due, in part, to the existence of the magnetoelastic coupling [6,7] and also due to the fact that transverse phonons can hold orbital momentum and consequently are a potential reservoir for spin-phonon interaction [4,7,8]. As a matter of fact, using phonons as an efficient channel to drive materials in new states becomes promising. In this poster, we will show how to generate and detect with visible light coherent acoustic phonons in multiferroics $BiFeO_3$ (LA, TA) [9-12] and, with time-resolved X-ray, how to quantitatively evaluate the ultrafast longitudinal and transverse light-induced strain [12].

References

[1] A. Kimel et al, Physics Reports 852, 1 (2020).
[2] C. Thomsen et al, Phys. Rev. B 34, 4129 (1986).
[3] P. Ruello, V. Gusev, Ultrasonics 56, 21-35 (2015).
[4] C. Dornes et al, Nature 565, 209 (2019).
[5] M. Deb et al, Phys. Rev. Lett., 123(2):027202.
[6] A. Stupakiewicz et al Nature Physics, *17*(4), 489-492.
[7] M. Lejman et al, Phys Rev B, 99, 104103 (2019).
[8] V. Juvé et al, Phys Rev B, 102, 202303 (2020)
[9] P. Ruello et al, App. Phys. Lett. 21, 212906 (2012).
[10] M. Lejman et al, Nat. Comm. 5, 4301 (2014).
[11] M. Lejman et al, Nat. Comm. 7, 12345 (2016)
[12] Gu et al., Sci. Adv. 9, eadi1160 (2023)
[15]A. Levchuk et al, Phys. Rev. B 101, 180102(R) (2020).
[16]R. Gu et al, to be submitted.

6

Generation of THz radiation via polariton parametric scattering in a rectangular $LiNbO_3$ waveguide

I.Betka [1*], M. Deroh[2], M. E. Ojo[1], F. Fauquet[1], C. Fourcade-Dutin[1], H. Maillotte[2], M. Chauvet[2], P. Mounaix[1], D. Bigourd[1 *]

1. Laboratoire IMS, UMR CNRS 5218, University of Bordeaux, 33400 Talence, France
2. Institut FEMTO-ST, Département d'Optique, UMR CNRS 6174 -Université Bourgogne Franche-Comté, 25030 Besançon, France

**Corresponding author: ilyes.betka@u-bordeaux.fr*

Polariton parametric scattering in polar crystals such as Lithium Niobate (LN) is a promising method to generate coherent THz radiations [1]. This process has been mostly investigated to deliver tuneable narrow-band THz radiation [2] when a long pulse pumps the material. A significant challenge hindering this technique lies in the generation of a THz spectrum centred at frequencies exceeding 2 THz, primarily due to the gain spectrum of LN, and the important absorption coefficient at higher frequencies [3]. In this submission , we highlight the possibility to overcome this limitation by pumping the LN in a transient regime. An Ytterbium based ultra-fast laser, delivering 400fs ultra-short pulse at a repetition rate of 85 kHz has been used to pump a 500 × 500 μm rectangular $LiNbO_3$ waveguide at room temperature. The length of the waveguide is 15 mm. Electro-optic detection have provided a comprehensive characterization of the temporal and spectral profiles of the generated THz wave. Figure 1 shows an example of a temporal trace recorded when the average power is set at 600 mW. The fast Fourier spectrum of the EO sampling measurement (Figure 2) shows a broadband peak with a width of ~ 4 THz (at full-width at half maximum) centred at 3 THz. Our results agree with the theoretical phonon polariton dispersion curve and the diffraction modified Schwarz-Maier plane wave model [4]. In this case, the mode area mismatch between the optical and the THz fields together with the unique waveguide structure which minimizes the diffraction-induced absorption lead to a modification in the gain spectrum, centred at 3 THz. Given the compatibility of waveguides with on-chip fabrication and their compact footprint, this research is a good step forward to develop an elegant platform for realizing on-chip THz radiation generation through nonlinear frequency conversion processes.

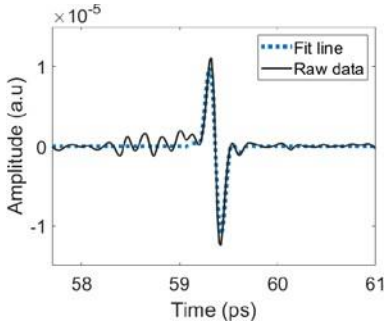


Figure 1 : Temporal wave form of the detected THz pulse

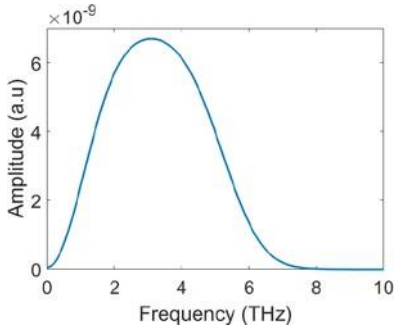


Figure 2 : Corresponding spectra obtained by FFT

References:

[1] M. A. Piestrup, R. N. Fleming, and R. H. Pantell, "Continuously tunable submillimeter wave source," Appl. Phys. Lett. 26, 418–421 (1975).

[2] A. M. Warrier, R. Li, J. Lin, A. J. Lee, H. M. Pask and D. J. Spence, "Tunable terahertz generation in the picosecond regime from the stimulated polariton scattering in a LiNbO 3 crystal." Optics Letters 41.18 ,4409-4412 (2016).

[3] U. T. Schwarz and M. Maier. "Frequency dependence of phonon-polariton damping in lithium niobate." Physical Review B 53.9, 5074 (1996).

[4] T. Wang, Y. Huang, M. Chuang, Y. Lin, C. Lee, Y. Lin, F. Lin and G. K. Kitaeva . "Long-range parametric amplification of THz wave with absorption loss exceeding parametric gain." Optics Express 21.2, 2452-2462 (2013).

7

Current status of deMon2k for the investigation of the early stagesof matter irradiation by time-dependent DFT approaches

Karwan A. Omar[1,2], Feven A. Korsaye[1,3], Rika Tandiana[1], Damien Tolu[1,4], Jean Deviers[1,5], Xiaojing Wu[6], Angela Parise[7], Aurelio Alvarez-Ibarra[1], Felix Moncada[8], Jesus Nain Pedroza-Montero[9], Daniel Mejia-Rodriguez[10], Nguyen-Thi Van-Oanh[1], Fabien Cailliez[1], Carine Clavaguéra[1], Karim Hasnaoui[11,12], Aurélien de la Lande[1]*

1. Université Paris Saclay, CNRS, Institut de Chimie Physique, UMR 8000, 91405, Orsay, France.
2. Department of Chemistry, College of Education, University of Sulaimani, 41005, Kurdistan, Iraq.
3. PSL University, CNRS, Chimie ParisTech-PSL, Institute of Chemistry for Life and Health Sciences (i-CLeHS), Theoretical Chemistry and Modelling Group (CTM), 75005 Paris, France.
4. CEA, DES, ISEC, DMRC, Université Montpellier, Marcoule, 30207 Bagnols-sur-cèze, France.
5. Department of Physics and Living Systems Institute, University of Exeter, Stocker Road, EX4 4QD Exeter, United Kingdom.
6. Department of Physics and Astronomy, University College London, London WC1E 6BT, United Kingdom.
7. Consiglio Nazionale delle ricerche/National Research Council (CNR)-IOM c/o International School for Advanced Studies (SISSA/ISAS), via Bonomea 265, 34136 Trieste, Italy.
8. Department of Physics, AlbaNova University Center, Stockholm University, S-106 91 Stockholm, Sweden
9. Institut Charles Gerhardt de Montpellier, CNRS, ENSCM, Université de Montpellier, 34296 Montpellier, France.
10. Environmental Molecular Sciences Laboratory, Pacific Northwest National Laboratory, Richland, WA 99352, USA
11. Institut du développement et des ressources en informatique scientifique, CNRS, UAR 851, bat. 506 BP 167 91403 ORSAY CEDEX – France
12. Maison de la Simulation, UAR 3441, Bâtiment 565 PC 190 – Digiteo CEA Saclay 91191 Gif-sur-Yvette cedex – France

* http://hebergement.universite-paris-saclay.fr/adelalandecnrs/

We summarize on this poster the recent progress made in our laboratories in the development of numerical approaches dedicated to investigating ultrafast physicochemical responses of biological matter subjected to ionizing radiations. Our modules are integrated into the deMon2k software which is a readily availableprogram with highly optimized algorithms for conducting Auxiliary-Density-Functional-Theory (ADFT) calculations. We have developed a computational framework based on Real-Time Time-dependent ADFT to simulate the electronic responses of molecular systems to strong perturbations, while molecular dynamicssimulations in the ground and excited states (Ehrenfest dynamics) are available to simulate irradiation-induced ultrafast bond breaking/formation[1].
Constrained ADFT and Multicomponent ADFT[2] have also been incorporated to simulate charge transfer processes and nuclear quantum effects, respectively. Finally, a coupling to polarizable force fields furtherpermits to realistically account for the electrostatic effects that the systems' environment has on the perturbed electron density. The code runs on CPU or hybrid CPU/GPU architectures affording simulations of systems comprised of up to 1000 atoms at the DFT level with controlled numerical accuracy[3]. Thecode can be downloaded on the Zenodo server (https://zenodo.org/records/10006417). Examples of recent applications will be shown on the poster.

References:

[1] Omar K A, Korsaye F A, Tandiana R, Tolu D, Deviers J, Wu X, Parise A, Alvarez-Ibarra A, Moncada F, Pedroza-Montero J N, Mejía-Rodriguez D, Van-Oanh N-T, Cailliez F, Clavaguéra C, Hasnaoui K and de la Lande A 2023 Current status of deMon2k for the investigation of the early stages of matter irradiation by time- dependent DFT approaches *The European Physical Journal Special Topics* **232** 2167–93

[2] Mejía-Rodríguez D and de la Lande A 2019 Multicomponent density functional theory with density fitting *J. Chem. Phys.* **150** 174115

[3] Martínez P A, Vock T, Kharchi L R, Pedroza-Montero J N, Wu X, Hasnaoui K and de la Lande A 2024 A multi-GPU implementation of Real-Time Time-Dependent Auxiliary Density Functional Theory for the investigation of nanosystems irradiations *Computer Physics Communications* **295** 108946

8

Photon pathways in the nonperturbative nonlinear regime of high harmonic generation

Titouan Gadeyne,[1] Mekha Vimal,[1] Martin Luttmann,[1] Matthieu Guer,[1] Romain Cazali,[1] Céline Chappuis,[1] David Bresteau,[1] Fabien Lepetit,[1] Olivier Tcherbakoff,[1] Jean-François Hergott,[1] Thierry Auguste,[1] and Thierry Ruchon[1]

[1]*Université Paris-Saclay, CEA, CNRS, LIDYL, 91191 Gif-sur-Yvette, France*

High harmonic generation (HHG), now a landmark of attosecond science, is best known as a strong-field process in which a driving laser wave interacts with the atoms of a gas target to produce radiation at high-order multiples ofits frequency. In parallel, the generation of a photon of harmonic order q can be seen as coming from the absorptionof q driving photons and subsequent emission of a single harmonic photon in the nonlinear medium. Though this photon-based reasoning correctly predicts the energy and momenta of high-harmonic photons, it does not account forthe strongly nonperturbative yield of HHG: from perturbative nonlinear optics, a q-photon process is expected to scaleas E^q, whereas plateau high harmonics all scale as $E^{q_{\text{eff}}}$ with a common effective nonlinearity order $q_{\text{eff}} < q$. In this talk, we will present a photon-based interpretation of HHG that correctly accounts for its nonperturbative efficiency, backed by experimental observations and a simple analytical model.

We will present experimental results on the yields [1], transverse modes [2] and phases of beamlets produced when driving HHG with two noncollinear beams. As demonstrated in recent years [3, 4], perturbative power laws are observed in this arrangement when the second beam is much weaker than the other. Investigating the regime where the two beams have comparable intensities, we will show the nonperturbative nature of HHG to be responsible for a wealth of unexpected observations, unseen in perturbative harmonic generation processes: oscillatory yield curves, transverse mode reshaping, among others. We will introduce an analytical framework explaining these features of nonperturbative nonlinear optics as originating from the coherent addition of infinitely many photon pathways involving absorption- emission pairs, with appropriate weights (see [1, 2] and Figure 1, bottom). Their interference is experimentally revealed through secondary maxima in yield curves (see [1] and Figure 1, top) and transverse mode coupling [2]. This first experimentally-backed extension of the photon picture of nonlinear optics to the strongly nonperturbative regime furthermotivates the search for a practical quantum-optical theory of HHG, and may find applications in spectroscopy and all-optical extreme-ultraviolet beam shaping.

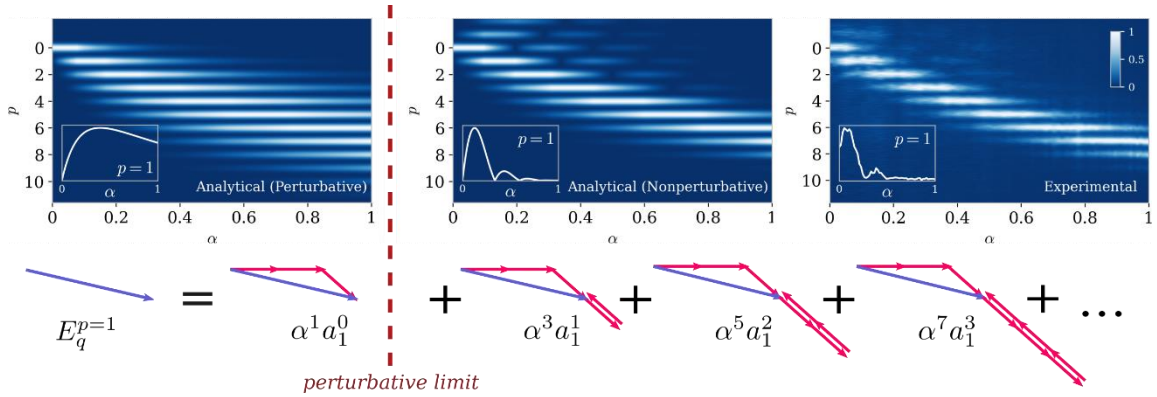


FIG. 1. **From perturbative to nonperturbative nonlinear optics.**
Top : contour plots of the yields of the beamlets (labelled p, vertical axis) from HHG driven by two non collinear beams, asa function of the amplitude ratio a between the driving beams. Beamlet $p = 0$ (resp. $p = q$, the harmonic order)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

[1] M. Vimal, M. Luttmann, T. Gadeyne, M. Guer, R. Cazali, D. Bresteau, F. Lepetit, O. Tcherbakoff, J.-F. Hergott, T. Auguste,and T. Ruchon, Physical Review Letters **131**, 203402 (2023).

[2] M. Luttmann, M. Vimal, M. Guer, T. Gadeyne, C. Chappuis, J.-F. Hergott, and T. Ruchon, Physical Review A **108**, 053509(2023).

[3] J. B. Bertrand, H. J. Worner, H.-C. Bandulet, É. Bisson, M. Spanner, J.-C. Kieffer, D. M. Villeneuve, and P. B. Corkum, Physical Review Letters **106**, 023001 (2011).

[4] L. Li, P. Lan, L. He, X. Zhu, J. Chen, and P. Lu, Physical Review Letters **120**, 223203 (2018)

9

Simulating attochemistry: How good are mixte quantum-classical approaches?

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

1. Nantes Université, CNRS, CEISAM UMR 6230, F-44000 Nantes, France

Since the advent of the first attosecond pulse in 2001,[1] its application on polyatomic moleculeshas given birth to the field of attochemistry. The large energy bandwidth of such laser source will coherently populate multiple electronic states i.e. a coherent electronic superposition. A particular challenge of this field is the accurate theoretical simulation of the molecular coupledelectron-nuclear dynamics induced by an electronic wavepacket.[2] It is particularly difficult forstandard mixte quantum-classical method to properly describe the electronic coherence and its effect on the subsequent nuclear dynamics. In our investigation on an example system withtwo cationic electronic states of 1-fluorobenzene where a conical intersection (CI) is located inthe vicinity of the Franck-Condon point (FC), we are assessing whether mixte quantum- classical methods such as Tully surface hopping and classical Ehrenfest are able to qualitatively reproduce the average motion of a quantum wavepacket propagated with Direct Dynamics variational Multiconfigurational Gaussian (DD-vMCG)[3] in the branching space.

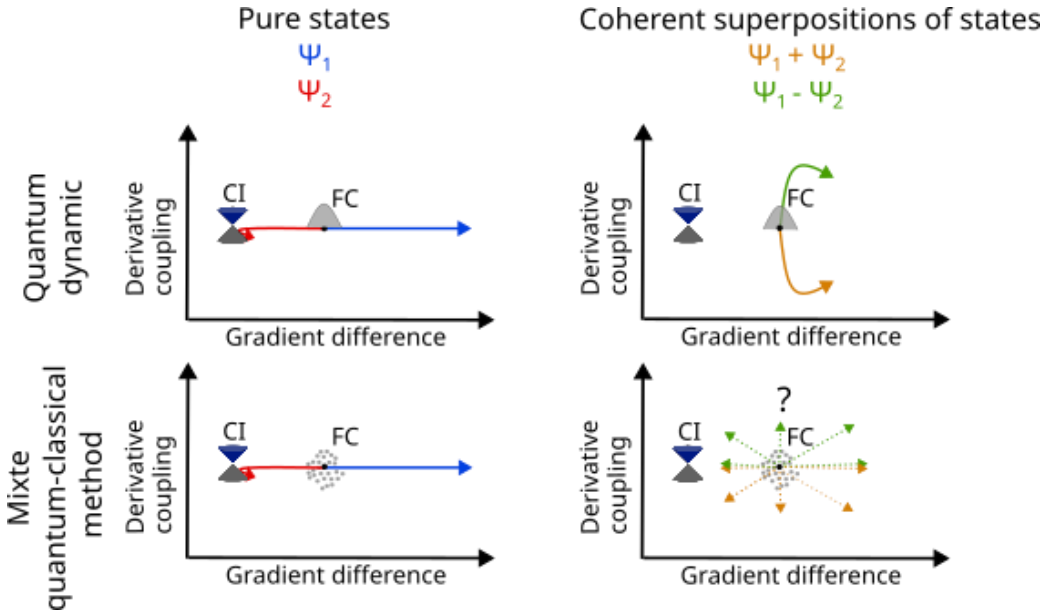


Figure: Scheme illustrating the average nuclear motion in the branching space of 1-fluorobenzene in the cationic manifold for a set of DD-vMCG (top) and mixte quantum-classical (bottom) dynamics initiated on a pure lower (Ψ_1) and upper (Ψ_2) states (left) and a coherent superposition with a different relative phase (right).

References:

[1] Paul, P. M.; Toma, E. S.; Breger, P.; Mullot, G.; Augé, F.; Balcou, P.; Muller, H. G.;Agostini, P. "Observation of a Train of Attosecond Pulses from High Harmonic Gener-ation" Science 2001, 292, 1689–1692.

[2] Cederbaum, L.; Zobeley, J. "Ultrafast charge migration by electron correlation", ChemicalPhysics Letters 1999, 307, 205 – 210.

[3] G. A. Worth and I. Burghardt, "Full quantum mechanical molecular dynamics using gaussianwavepackets", Chemical Physics Letters 2003, 368, 502 – 508.

10

Ultra-short Laser-Induced Modifications and Properties of Magneto-Optical Nanoparticles

Tatiana E. Itina[1], Mohammad Sadegh Shakeri[2], Oliwia Polit[2], Andrei Pastukhov[3], Andrei Kabashin[3], Zaneta Swiatkowska-Warkocka[2]

[1]Université Jean Monnet Saint-Étienne, CNRS, Institut d'Optique Graduate School, LaboratoireHubert Curien UMR 5516, F-42023, Saint-Étienne, France

[2]Institute of Nuclear Physics Polish Academy of Sciences, PL-31342 Krakow, Poland

[3]Université Aix-Marseille, CNRS, Laboratoire Lasers Plasmas et Procédés Photoniques, UMR 7341,

Marseille, France

tatiana.itina@univ-st-etienne.fr

Short and ultra-short laser interactions in liquids open numerous promising possibilities for the modifications of colloidal nanoparticles. In particular, femtosecond laser irradiation of colloidal nanoparticles can either lead to their ablation and fragmentation or to their sintering, or, more generally, to both of these processes. In addition, such experiments allow the mixing of normally non-miscible compounds or the creating of complex multi-core-shells and/or core- satellite nanoparticles that may have particularly interesting combined magneto-optical properties.

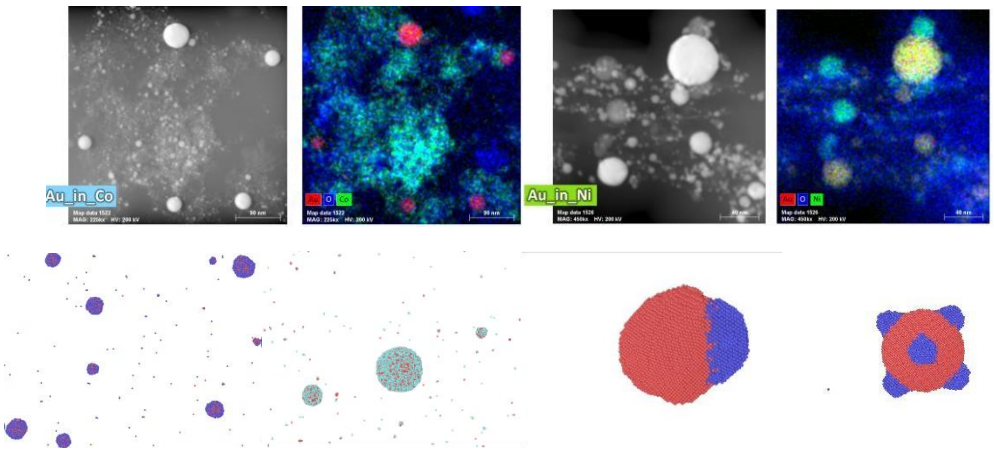


Fig. 1. Experimental and numerical (MD) results obtained for AuCo, and AuNi nanoobjects under several conditions.

In this collaborative study, both atomistic molecular dynamics (MD) simulations and several series of experimental studies are performed with several nanoparticles composed of both plasmonic and magnetic-optical combinations, such as Au-Co and Au-Ni. In particular, the difference in sizes, initial composition, alloying with Au, binding and surface energies as wellas in the ductile properties are shown to affect the final configurations and the resulting magnetic and optical properties

References

[1] D. Zhang, B. Gokce, and S. Barcikowski. Laser synthesis and processing of colloids: fundamentalsand applications. *Chemical reviews*, *117*(5), 3990-4103 (2017).

11

Attosecond transient absorption in a strong field

Léonardo Rico[1], Juliette Dubois[1,2], Jérémie Caillat[1], Boris Vodungbo[1], Richard Taïeb[1], Guillaume Lambert[2], Camille Lévêque[1]

1. Sorbonne Université, CNRS, LCPMR, 4 place Jussieu, 75005 Paris, France
2. LOA, ENSTA Paris, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, 828 Bd des Maréchaux, 91762 Palaiseau, France

Transient absorption is a typical time-resolved pump-probe experimental scheme that was first used to probe millisecond timescale dynamics [1]. As the laser pulses got shorter and more intense, this technic became able to probe faster phenomena. In particular, the use of High Harmonic Generation (HHG)
[2] gives access to the attosecond timescale, which is the natural scale of electrons dynamics in matter. Typical attosecond transient absorption experiments use an extreme UV (XUV) pulse as a pump and either another XUV or an IR pulse as a probe [3]. In the case of an IR probe pulse, its intensity is usually of the order of 10^{12} W.cm^{-2}.

In this work, we study transient absorption of various atomic species in the gas phase using an unusually intense (10^{14} W.cm^{-2}) IR pulse (pump) and an attosecond train pulse (probe), motivated by experiments done at Laboratoire d'Optique Appliquée. We present and analyze results of numerical simulations obtained in the single-active electron approximation in 2D [4,5] to interpret this scheme as a new way to measure IR pulse durations.

References:

[1] 1967 Chemistry Nobel prize decerned to Manfred Eigen, Ronald G. W. Norrish and George Porter

[2] 2023 Physics Nobel prize decerned to Pierre Agostini, Ferenc Krausz and Anne L'Huilier

[3] Mengxi Wu et al 2016 J. Phys. B: At. Mol. Opt. Phys. 49 062003

[4] André D. Bandrauk, Hai Shen; J. Chem. Phys. 15 July 1993; 99 (2): 1185-1193

[5] J. Caillat, A. Maquet, F. Risoud, R. Taïeb, in Attosecond Molecular Dynamics, ed. M. J. J. Vrakking and F. Lepine, The Royal Society of Chemistry, 2018, pp 38-67.

[6] A. Zaïr et al, Appl. Phys. B 78, 869-872 (2004)

12

Terahertz-driven field evaporation: application to Atom Probe Tomography

M. Karam[1], J. Houard[1], G. Darmala[1], S. Idlahcen[2],A. Martinez[3], D. Paparo[3,4], A. Hideur[2], A. Vella[1]

1. Univ Rouen Normandie, INSA Rouen, CNRS, Groupe de Physique des Matériaux, 76000 Rouen, France
2. CORIA CNRS, INSA, Université de Rouen Normandie, 76801 Saint Etienne du Rouvray, France
3. Dipartimento di Fisica 'E. Pancini', Università 'Federico II', Monte S. Angelo, via Cintia, 80126 Napoli, Italy
4. CNR-ISASI, via Campi Flegrei 34, Pozzuoli, Italy

Terahertz (THz) monocycles are useful for various applications, including electron emission, surface tunneling microscopy (STM) and atom probe tomography (ATP). APT is a powerful nanoscale imaging and analysis technique used to characterize the composition and spatial distribution of atoms within materials. It is based on atom-by-atom ionization, ejection and detection, allowing for three-dimensional reconstruction of the sample's atomic structure, by a projection-back law. APT provides the chemical information on each ion by time-of-flight mass spectrometry. Recently we have shown that single-cycle THz pulses can be used to ionize and eject positive ions from an APT sample, biased at a positive voltage of several kilovolts [1].

In this contribution, we report on the use of mono-cycle THz pulses to emit electrons or ions from LaB_6 samples. The THz generation bench is based on two-color laser air-plasma is exploited (Fig. 1). The terahertz radiation can be alternatively focused on the electro-optics (EO) sampling setup based on a GaP crystal or on the nanotip inside the atom probe chamber. By changing the applied bias from negative to positive values we can emit electrons or the ions from the sample. By electron emission we are able to measure the THz near field at the apex of the nanoneedle. Looking at the time- of-flight (TOF) spectra of emitted ions, we have information on their acceleration and/or deceleration by the THz pulse. We use a commercial particle trajectory analysis software LORENTZ-2E V10.2 to calculate the straight flight path of ions in the atom probe chamber under the action of the static electric field and the THz transient. Numerical simulations are in good agreement with the experimental results.

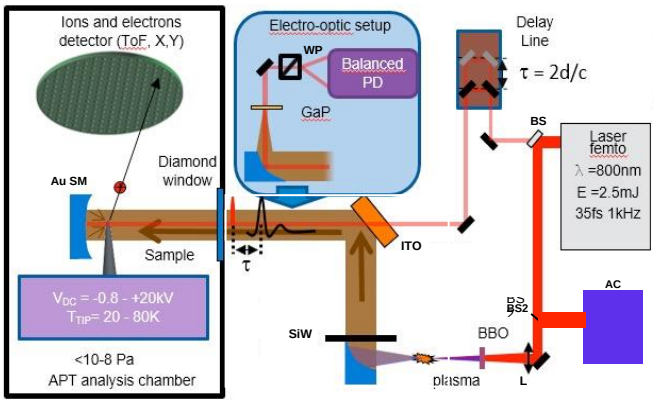


Fig. 1 Experimental setup of THz-assisted atom probe

References:

[1] Vella, A., Houard, J., Arnoldi, L., Tang, M., Boudant, M., Ayoub, A., ... & Hideur, A. (2021). High-resolution terahertz-driven atom probe tomography. *Science Advances*, *7*(7), eabd7259

[2] Houard, J., Arnoldi, L., Ayoub, A., Hideur, A., & Vella, A. (2020). Nanotip response to monocycle terahertz pulses. *Applied Physics Letters*, *117*(15), 151105.

[3] Karam, M, Houard, J, Damarla,G., Rousseau, L., Bhorade, O.,Vella, A (2021). THz driven field emission: energy and time-of-flight spectra of ions. *2023 New J. Phys. in press* https://doi.org/10.1088/1367-2630/ad0855

13

Ultrafast Femtosecond Laser Spectroscopy of Heterogeneous SolidPhotoactive Catalysts for The Conversion of Methane

Nour Alhajjar[1], Thomas Roland[1], Yinghao Wang[2], Andreï Khodakov[2], Vincent De Waele[1]

1. LASIRe – Laboratoire Avancé de Spectroscopie pour les Intéractions la Réactivité et l'Environnement, Univ. Lille, CNRS, UMR 8516, F-59000 Lille, France

2. UCCS – Unité de Catalyse et Chimie du Solide, Université de Lille, CNRS, Centrale Lille, ENSCL, Université d'Artois, UMR 8181, F-59000 Lille, France

Methane (CH_4) holds great potential as a precursor for the synthesis of value-added chemicals and sustainable fuels, crucial for achieving a decarbonized industry. However, its inherently stable C-H bonds necessitate high-temperature activation, resulting in significant energy costs and limited selectivity. Therefore, the development of efficient and selective photocatalytic routes for methane conversion is necessary. Whereas it is well accepted that the photocatalysis is triggered by the formation of electron-hole pairs in TiO_2, the link between the ultrafast photodynamics and the efficiency of the catalyst is not established. To fill this gap, this project aims to carry out femtosecond time-resolved spectroscopy to unravel the primary chemical processes triggering the photo-conversion of methane and to establish clear relationships between these ultrafast processes and the catalytic activity. Our focus centers on innovative photocatalysts based on TiO_2 [1] with the generic formula (Pt/NPW)/TiO_2, a ternary-phase composite consisting of a TiO_2 support and ammonium phosphotungstic polyoxometalate (NPW) sub-nanoclusters anchored with isolated Pt single atoms (Pt_1) [2].

We are reporting on transient absorption spectra measurements in the mid-infrared (MIR) region at a wavelength of 4800 nm, specifically targeting the spectral region where only free electrons within the conduction band or shallow-trapped electrons absorb. All the measurements were performed on self-supported pellets in KBr under controlled atmosphere condition. We investigated the impact of (Pt/NPW) concentration and pump intensity on the temporal evolution and concentration of photogenerated charge carriers within the catalysts.

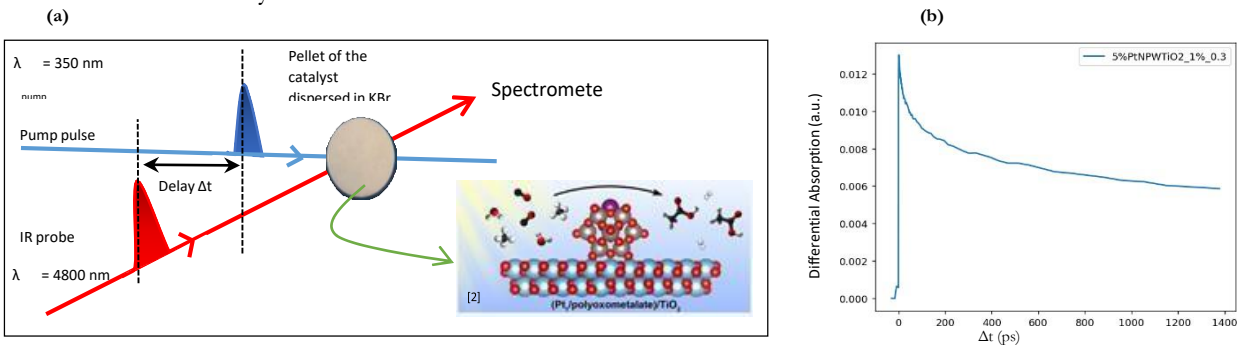


Fig.1 (a) Scheme of pump-probe measurements on (Pt/NPW)/TiO_2 pellet. (b) Kinetic trace at 4.8 micrometer obtained for 5%(Pt/NPW)/TiO_2 with λ_{pump} = 350 nm.

References:

[1] X. Yu et al., Nature Communications 10(1), 700 (2019).
[2] Dong, C., et al., Journal of the American Chemical Society, 145(2), 1185-1193 (2023).

Although it was observed almost 30 years ago, the dynamics of magnetization at ultimate femtosecond and attosecond time scale remains a hot topic. In part, this is due to the lack of dedicated and suitable experimental insights. While most of ultrafast measurements have been carried out with visible lasers, progress of attosecond science, which offers ultrashort light pulses in the eXtreme UltraViolet spectral range, let envision radically new approaches. In particular, contrary to visible light, it is suitable to address specific edges of many magnetic materials in magnetic alloys and heterostructures. However, up to recent days, control of its angular momenta was difficult, if possible at all.

The situation has changed lately, with the demonstration of reliable attosecond light sources carrying either spin or orbital angular momentum. When interacting with magnetic matter, SAM can be modified through magnetic circular dichroism (MCD). As for OAM, we recently showed that the beam's content depends on the material's spin texture upon reflection, a process that we called magnetic helicoidal dichroism (MHD). Here we present recent developments in building a high harmonic generation-based beamline dedicated to the study of both MCD and MHD in solids with attosecond resolution in a transient spectroscopy scheme. First commissioning experiments were performed on gaseous and solid samples with linear polarization, showing sub-cycle resolution. On a solid MgO sample, the attosecond dynamic of multiple core-excitons have been measured, showing a complex behavior explained by the dynamical Franz-Keldysh effect. Finally, our efforts towards the more difficult task of magnetic dichroisms will be presented.

15

Ultrafast Exciton Dynamics in 2D van der Waals nanostructures: Probing the Hot Exciton Relaxation of Size-Controlled & Well Dispersed Graphene Nanoflakes

Sébastien QUISTREBERT[1], Daniel MEDINA-LOPEZ[1], Stéphane CAMPIDELLI[2], Jean-Sébastien LAURET[1], Elsa CASSETTE[1]

1. LuMIn (UMR 9024), Université Paris-Saclay, ENS Paris-Saclay, CNRS, CentraleSupélec, 91400 Orsay, France
2. NIMBE/LICSEN (UMR 3685), Université Paris-Saclay, CEA, CNRS, 91191 Gif-sur-Yvette, France

Graphene nanostructures, such as graphene quantum dots (G-QDs), graphene nanoribbons (G-NRs) and carbon nanotubes (C-NTs), combine the unique mechanical and electronical transport properties of sp^2- hybridized carbon materials and the optical properties of direct semiconductors provided by the optical gap resulting from the reduction of dimensionally. Among them, the recent developments within the well-known synthesis of G-QDs though bottom-up approach [1] have led to exceptionally well-controlled nanostructures in terms of size, shape and dispersion [2]. The resulting graphene nanoflakes provide tunable emission in the red range, with fluorescence quantum yield close to 1. Furthermore, these nanostructures have revealed to be promising stable emitters of single photons, as shown in our laboratory [2-5].

Here we use transient absorption of 30 fs temporal resolution with polarization-controlled configuration to probe the hot exciton relaxation (internal conversion, $S_n \rightarrow S_1$) in rectangular G-QDs of various lateral lengths. The nanoflakes are composed of exactly 96, 114 and 132 conjugated carbons (respectively 2.30, 2.71 and 3.11 nm). While the ultrafast electronic dynamics in graphene nanostructures are often being blurred by large broadband photo-induced absorption signals [6-8] (in particular involving triplet states, $T_1 \rightarrow T_n$), here the suppressed aggregation in the studied graphene nanoflakes allows a clear observation and identification of the discrete ground state bleaching and photo-induced emission signals.

We selectively excite the different samples at the second optically active electronic transition and, thought the appearance of a photo-induced emission signal at the energy corresponding to the bandedge and red-shifted vibrational replica (*i.e.* at the position of the steady-state photoluminescence peaks), the dynamics of relaxation were unveiled. The resulting relaxation times range from 100 fs to 175 fs. These results allowed to discuss the mechanism of relaxation, with the effect of the length of the graphene nanoflakes and of the fluence excitation [Quistrebert *et al.*, in preparation].

References:
[1] A. Narita, X.-Y. Wang, X. Feng, K. Müllen. Chem. Soc. Rev. 44, 6616 (2015).
[2] D. Medina-Lopez, T. Liu, S. Osella, H. Levy-Falk, N. Rolland, C. Elias, G. Huber, P. Ticku, L. Rondin, B. Jousselme, D. Beljonne, J.-S. Lauret, S. Campidelli. Nat. Commun. 14:4728 (2023).
[3] T. Liu, B. Carles, C. Elias, C. Tonnelé, D. Medina-Lopez, A. Narita, Y. Chassagneux, C. Voisin, D. Beljonne, S. Campidelli, L. Rondin, J.-S. Lauret. J. Chem. Phys. 156, 104302 (2022)
[4] T. Liu, C. Tonnelé, S. Zhao, L. Rondin, C. Elias, D. Medina-Lopez, H. Okuno, A. Narita, Y. Chassagneux, C. Voisin, S. Campidelli, D. Beljonne, J.-S. Lauret. Nanoscale 14, 3826 (2022).
[5] S. Zhao, J. Lavie, L. Rondin, L. Orcin-Chaix, C. Diederichs, P. Roussignol, Y. Chassagneux, C. Voisin, K. Müllen, A. Narita, S. Campidelli, J.-S. Lauret. Nat. Commun. 9 :3470 (2018).
[6] M.L. Mueller, X. Yan, B. Dragnea, L.s. Li. Nano Lett. 11, 56-60 (2011).
[7] D. Sebastian, A. Pallikkara, H. Bhatt, H.N. Ghosh, K. Ramakrishnan. J. Phys. Chem. C 126, 11182-11192 (2022).
[8] M. Reale, A. Sciortino, M. Cannas, E. Maçoas, A.H.G. David, C.M. Cruz, A.G. Campana, F. Messina. Materials 16, 835 (2023).

16

High harmonic generation in ZnO driven by a high energy fiber laser system

Djamila Boukhaoui[1], Said Idlahcen[2], Jonathan Houard[1], Ivan Blum[1], Thomas Godin[2], Foued Amrani[3], Frédéric Gérôme[3,4], Fetah Benabid[3,4], Angela Vella[1] and Ammar Hideur[2]

1. GPM, CNRS UMR 6634, INSA - University of Rouen, Normandie University, 76801 St.Etienne du Rouvray, France
2. CORIA, CNRS UMR 6614, INSA - University of Rouen, Normandie University, 76801 St.Etienne du Rouvray, France
3. Glophotonics, 123 avenue Albert Thomas, 87060 Limoges Cedex, France
4. GPPMM Group, XLIM Research Institute, CNRS UMR 7252, University of Limoges, 87060 Limoges, France

Abstract

Experimental studies on High harmonic generation from bulk crystal targets using longer IR driving laser pulses have yielded significant insights. These investigations revealed enhanced harmonic intensities in the XUV spectral range, an extended cut-off and a higher damage threshold for bulk crystal. This was achieved using an OPCPA [1- 4] and an OPA [5-10]. While offering a high energy levels, these systems present a very complex configuration. The use of ultrafast fiber lasers in the mid-IR emerges as a promising route for developing ultra-compact HHG platforms. This has been confirmed by several demonstrations of HHG driven by few-cycle mid-IR fiber lasers [11,12]. However, these sources based on soliton self-frequency shift in all-solid fibers are limited to few 10 nJ [11,12]

In our contribution, We report on high harmonic generation in solids driven by a high energy fiber laser system operating around 1550 nm. The driving laser source consists in an erbium-doped fiber chirped pulse amplifier combined with a post- compression stage featuring a hollow-core photonic crystal fiber (HC-PCF) filled with noble gases. The nonlinear self- compression occurring in the HC-PCF enable the generation of ultrashort pulses with 50 fs duration and 0.91 μJ at 660 kHz repetition rate at gas pressure higher than 11 bar. Focusing the compressed pulses into 500μm ZnO thick, we observe the generation of ultraviolet harmonics extending well above the band gap (Δ_{gap}=3.3 eV) and confirm the nonperturbative scaling for near- and above- threshold harmonics, we notice also the enhancement of the luminescence generated at the excitonic band edge (385 nm). In this experiment, we were limited to the measurement of H7 due to our detection system.

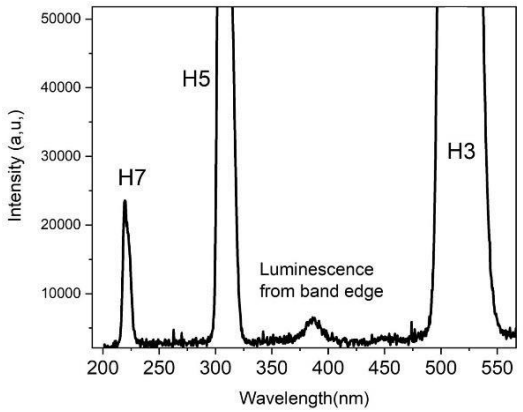


Fig: Spectra emitted from the ZnO measured at pump intensity of 3 TW/cm²

References

[1] S. Ghimire et al. Nat. Phys 7, 138, (2011)
[2] T. Kroh et al. Opt. express 26.13 :16955-16969 (2018)
[3] H. Richard, et al. Opt. Express 28, 7314 (2020)
[4] V. E. Nefedova, et al. Appl. Phys. Lett 118, 201103 (2021)
[5] G. Mirzaei, Applied Physics Letters 110.6 (2017)
[6] A. A Lanin et al. Optica 4, 516-519 (2017)
[7] R. Hollinger et al. Nanomaterials 11, 4 (2020)
[8] S. Gholam-Mirzaei et al. JOSA B 35, A27 (2018)
[9] L. He et al. Ultrafast Science 3, 0022 (2023)
[10] K. Werner et al. Opt. Express 27, 2867-2885 (2019)
[11] D. Franz, et al. Sci. Rep 9, 5663 (2019)
[12] Y. Leventoux et al. Fiber Lasers XX: Technology and Systems. 12400. SPIE, (2023)

17

TDDFT simulations applied to pump-probe experiments with sapphire

K. Lévêque-Simon[1], R. Sinha-Roy[2], V. V´eniard1,

P. Lample[4], M. Weis[3], S. Guizard[4]

1. Laboratoire des Solides Irradiés (UMR 7642), CEA, CNRS, Ecole Polytechnique, 91128 Palaiseau Cedex, France
2. Institut Lumière Matière (UMR 5306), CNRS, Université Lyon 1, 69622 Villeurbanne Cedex, France
3. Laboratoire d'Optique Appliquée (UMR 7639), CNRS, Ecole Polytechnique, ENSTA-Paris, 91762 Palaiseau Cedex, France
4. Laboratoire Interactions, Dynamiques et Lasers (UMR 9222), CEA, CNRS, Université Paris Saclay, F-91191 Gif-sur-Yvette Cedex, France

The aim of this theoretical research work is to investigate the dynamics of solids in order to better understand the excitation and relaxation pro- cesses at ultrafast time scales. In particular, we will focus on wide band-gap materials like quartz (SiO_2) and sapphire (Al_2O_3) with optical cycle tem-poral resolution and beyond, with sub-femtosecond resolution. Thus we will address the physical mechanisms of laser-matter interaction and initial electronic relaxation in solids (multiphoton ionization, band gap modula- tion. . .) in connection with time resolved pump-probe experiments which currently operates at LIDYL/LOA [1].
We were first interested in the absorption spectrum for Al_2O_3 in the linear regime using the versatile *Octopus* DFT/TDDFT open-source software [2,3] based on a space/time resolution of the Kohn-Sham equations. In this approach, the dielectric function is obtained as the Fourier transform of the total electric current. We then carried out benchmarking of this spectra by considering as a reference those generated from the TDDFT linear response (DP) [4] and from the Bethe-Salpeter equation (EXC) [5] codes, which operate in reciprocal space and frequency domain. From this analysis, an estimate of the scissor correction leading to an optical bandgap around 9 eV has been obtained. In the future, this correction will improve our linear response spectra.
In the non-linear regime, we will present preliminary results for the change in reflectivity of the material after excitation by the pump. These results will be presented for several delays between the pump and the probe.

References:

[1] A. Bildé *et al.*, Journal of Physics: Condensed Matter **33**, 315402 (2021).
[2] N. Tancogne-Dejean *et al.*, J. Chem. Phys. **152**, 124119 (2020).
[3] X. Andrade *et al.*, Journal of Physics: Condensed Matter **24**, 233202 (2012).
[4] V. Olevano, L. Reining, and F. Sottile, http://dp-code.org.
[5] F. Sottile *et al.*, http://www.bethe-salpeter.org.

18

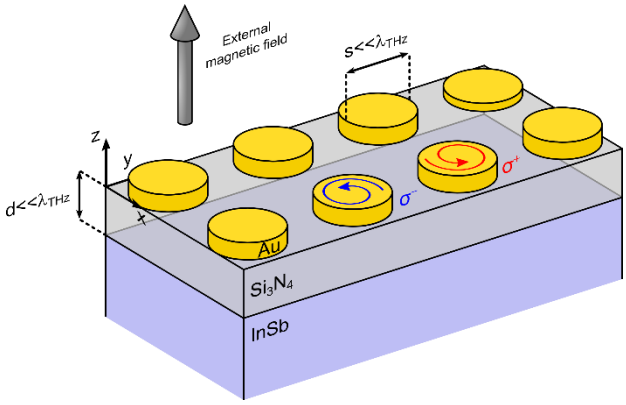
Chiral Terahertz surface plasmonic cavities

Ian Aupiais[1], Romain Grasset[1], Dmitri Daineka[2], Javier Briatico[3], Luca Perfetti[1], Jean-Paul Hugonin[4]

Jean-Jacques Greffet[4] and Yannis Laplace[1]

1. LSI/CEA/DRF/IRAMIS, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, Palaiseau, France Univ2
2. LPICM, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, Palaiseau, France
3. Unité Mixte de Physique, CNRS, Thales, Université Paris Saclay, 91767 Palaiseau, France
4. Université Paris-Saclay, Institut d'Optique Graduate School, CNRS Laboratoire Charles Fabry, 91127 Palaiseau, France

Ultrastrong light matter coupling can lead to hybrid light-matter states, opening new ways to manipulate material properties. In this context, Terahertz is particularly interesting because of the variety of excitations present in this low energy range. In this respect, the possibility to combine low energy chiral excitations with cavities having small mode volumes and chiral optical states is emerging as a new frontier for material manipulation [1]. Recently, we demonstrated ultrasmall and temperature tunable THz plasmonic cavities based on localized surface plasmon modes in semiconductor InSb [2]. Going one step further, here we demonstrate in these cavities ultrasmall mode volumes exhibiting chirality, meaning non-degenerate states for opposite circular polarizations. These results add new functionalities to these semiconductor based plasmonic cavities.



Schematic representation of chiral THz surface plasmonic cavities

References:

[1] H. Hübener et al. Nat. Mat. 20, 438–442 (2021)

[2] I. Aupiais et al. Nat. Comm. 14, 7645 (2023)

19

Conservation laws in photoionization by circularly polarized lights

Jonathan Dubois[1], Camille Lévêque[1], Jérémie Caillat[1], Richard Taïeb[1], Ulf Saalmann[2], Jan-Michael Rost[2]

1. Sorbonne Université, CNRS, Laboratoire de Chimie Physique Matière et Rayonnement, LCPMR, 75005 Paris, France
2. Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Straße 38, 01187 Dresden, Germany

We consider the process of tunnel ionization of atoms and molecules driven by circularly polarized (CP) pulses [1]. An intuitive semiclassical picture of tunnel ionization by a static laser field is an electron ionizing through the potential barrier induced by the laser field with constant energy, referred to as adiabatic ionization. When the laser field alternates in time, such as it is the case for CP pulses, the energyof the electron changes in time, and at the tunnel exit, it is distributed in a range of energy on the orderof the ponderomotive energy of the laser. The adiabatic picture no longer holds, and the ionization process is referred to as nonadiabatic [2]. Extensive theoretical and experimental studies are performed in the attosecond community to probe and understand these nonadiabatic effects in photoionization.

Our goal is to understand nonadiabatic processes in CP pulses using a semiclassical approach [3] in the combined laser and Coulomb fields. We map the electron dynamics in a frame that rotates with the laserfield, referred to as the rotating frame (RF). Our results show that in the RF, counter-intuitively, the energy of the electron is constant during tunnel ionization, and as a consequence follows the picture of adiabaticionization. This allows us to understand and predict, for instance, the role played by ring currents [4] inatoms and the shape of the laser envelope, and to shed light on classical-quantum correspondence.

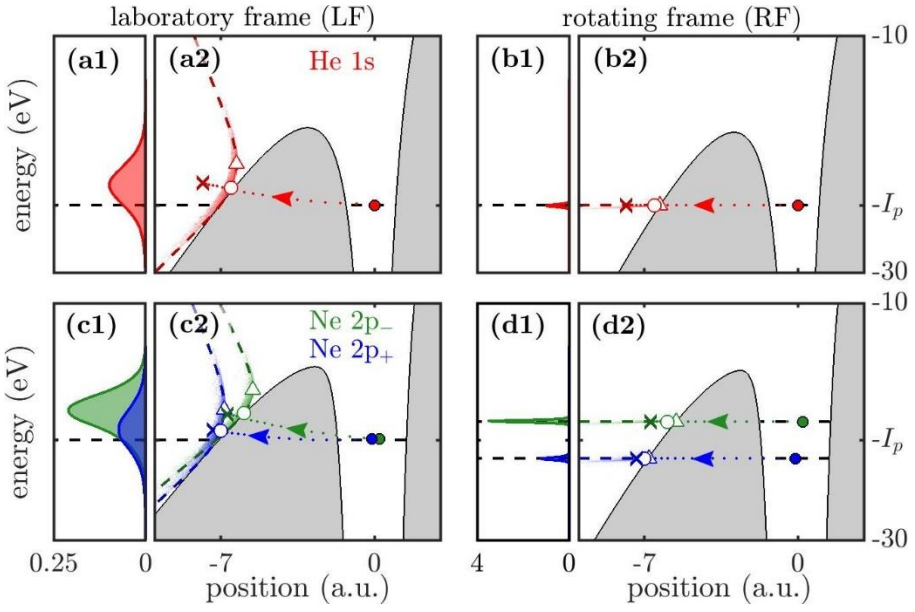


Figure 1. Configuration of the electron after tunnel ionization obtained by the backpropagation method [3]. (a) and (c) are the distributions in the LF. (b) and (d) distributions in the RF. Left panels are the distribution of the energyafter tunneling, right panels are the distribution in energy and in position along the field direction after tunneling.The grey regions indicate the classically forbidden region of the electron in the LF and in the RF at the peak amplitude of the laser field. The dotted curves are the electron complex trajectories under the classically forbiddenregion.

References

[1] Barth I *et al* 2011 *Phys. Rev. A* **84 063415**
[2] Boge R *et al* 2013 *Phys. Rev. Lett.* **111 103003**
[3] Ni H *et al* 2016 *Phys. Rev. Lett.* **117 023002**
[4] Eckart S *et al* 2018 *Nature Phys.* **14 701**

20

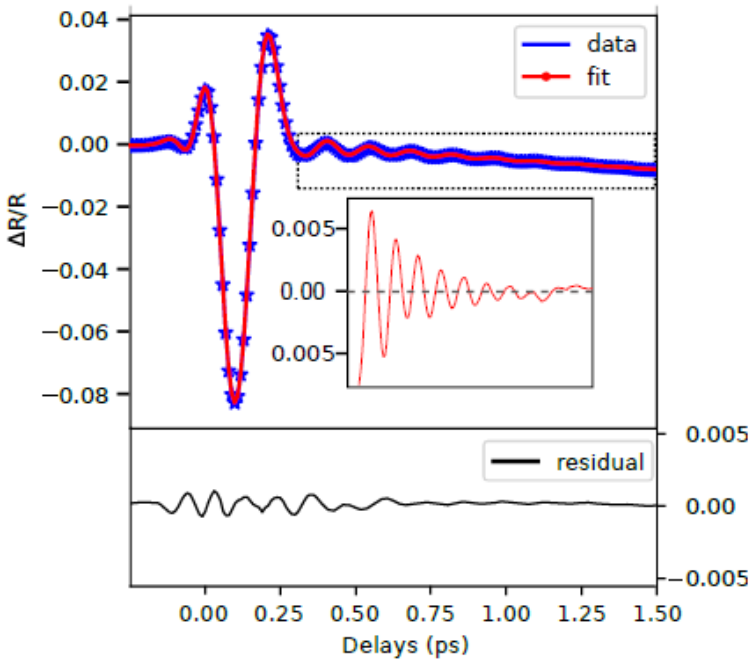
Coherent structural dynamics in V_2O_3 under hydrostaticpressure

Gauthier Thomas[1], Nicolas Godin[1] and Bertoni Roman[1], Janod Etienne[2], Corraze Benoit[2], Tranchant Julien[2], Danylo Babich[2] and Cario Laurent[2]

1. Univ Rennes, CNRS, IPR (Institut de Physique de Rennes) - UMR 6251, F-35000 Rennes, France
2. Nantes Université, CNRS, Institut des Matériaux de Nantes Jean Rouxel, IMN, Nantes, F-44322, Franc

Recently, material science has moved from observing and understanding electronic and structural orders to control physical properties of materials on command by usingexternal stimuli such as light. From this point of view, a crucial aspect is the understanding of the fundamental interactions within the system (electron-electron and electron-phonon) to control the emergent cooperative effects.

Here, we combine ultrafast optical spectroscopy and high-pressure setup to monitor the out-of-equilibrium dynamics of the material under well-defined controlled thermodynamical environment. The obtained results demonstrate the use of spectroscopy of coherent phonon as a thermodynamical phase marker of the Insulator to Metal Transition in V2O3 thinfilm. More intriguing, the variation of frequency of the observed phonon optical mode (A1g) seems to reflect the manifestation of critical coupling between lattice and electronic degrees of freedom near transition line with a drop of frequency near the critical pressure.



Transient reflectivity of V2O3 thin film around 675 nm after photo-excitation with 800 nm pulse at 6200 bars

References:

[1] Lantz, G., Mansart, B., Grieger, D. *et al.* Ultrafast evolution and transient phases of a prototype out-of-equilibrium Mott–Hubbard material. *Nat Commun* 8, 13917 (2017)

[2] S. R. Hassan, A. Georges, and H. R. Krishnamurthy,Phys. Rev. Lett. 94, 036402 – Published 24 January 2005

[3] M. Maesato et al, J. of Phys.: Conference Series148 (1), 012004, 2009

[4] Ultrafast photo-induced dynamics of V2O3 thin films under hydrostatic pressure G Privault, G Huitric, M Hervé, E Trzop, J Tranchant… - The European Physical Journal Special Topics, 2022

21

Spatial transformations of high-order harmonic generation in transition-metal dichalcogenides

Leon Schlemmer[1,2], Peng Ye[1], David Gauthier[1,2], Vijay Sunuganty[1,2], Sergey Babenkov[1,2], Marie Froidevaux[3], Xu Liu[1,2], Hamed Merdji[3], Willem Boutu[1,2]

1. Université Paris-Saclay, CEA, LIDYL, 91191 Gif sur Yvette, France

2. CY Cergy Paris Université, CEA, LIDYL, 91191 Gif sur Yvette, France

3. LOA, ENSTA ParisTech, CNRS, Ecole Polytechnique, 91120 Palaiseau, France

Since the first observation of high-order harmonic generation (HHG) from bulk crystals by S.Ghimire *et al.* in 2011, HHG in solids has been a growing field in ultrafast optics and condensed matter physics. In this work, we use the symmetry induced properties of the high-order harmonic emission from transition metal dichalcogenide (TMD) monolayers to locally influence the polarization and phase of the emitted harmonics. High-order harmonics were generated from mono- and polycrystaline molybdenum disulfide (MoS2) monolayers with an infrared femtosecond pulse. We control the orbital angular momentum (OAM) and spin angular momentum (SAM) of the generation beam by using a liquid crystal Q-plate. We then measure the OAM and the full polarization map of the emitted harmonics. On one hand we observe that, monocrystaline MoS2 monolayers behave as a polarization converter on even harmonics (4th harmonic), while they conserve the generation beam's OAM. On the other hand, we show that polycrystaline MoS2 monolayers can behave as a phase mask, which effectively modifies the OAM of the generated beam. The crystal symmetries only permit this remarkable behavior for even harmonics (4th harmonic). This work illustrates the potential of HHG from solids as a means to use the generating crystal's symmetries to locally tailor the harmonics polarization and phase.

22

Photoionization in a thick-lens VMIS with a HHG source

Nans Hermellin[1,2], Gabriele Crippa[2,3], Charles Bourrassin-Bouchet[4], Denis Cubaynes[2], Laila Dakroub[2], Annie Klisnick[2], Lionel Poisson[2], Lou Barreau[2]

1. Ecole Normale Supérieure Paris-Saclay, Université Paris-Saclay, Gif-sur-Yvette, France
2. Université Paris-Saclay, CNRS, Institut des Sciences Moléculaires d'Orsay, UMR 8214, 91405 Orsay, France
3. Université Paris-Saclay, CEA, CNRS, LIDYL, UMR 9222, 91191 Gif-sur-Yvette, France
4. Université Paris-Saclay, CNRS, IOGS, Laboratoire Charles Fabry, UMR 8501, 91120 Palaiseau

We present the implementation of a femtosecond photoionization spectroscopy setup using pulses in the extreme ultraviolet (XUV) region and a thick-lens Velocity Map Imaging Spectrometer (VMIS) for photoelectron detection [1,2]. The XUV probe beam is an attosecond pulse train generated from a femtosecond laser with a central wavelength of 800 nm through high-order harmonic generation (HHG). Various parameters are tested to optimize the generation, including the focusing conditions, the choice of gas, the pressure, and the geometry of the generation cell. Using Argon, it is possible to reach the 35^{th} harmonic (54 eV) and with Helium the 39^{th} harmonic (60 eV). This radiation is used to calibrate and determine the resolution of an 11-electrode VMIS, found to bebelow 5% for electron kinetic energies in the 4-16 eV range (see Figure 1), in good agreement with *SIMION* simulations.

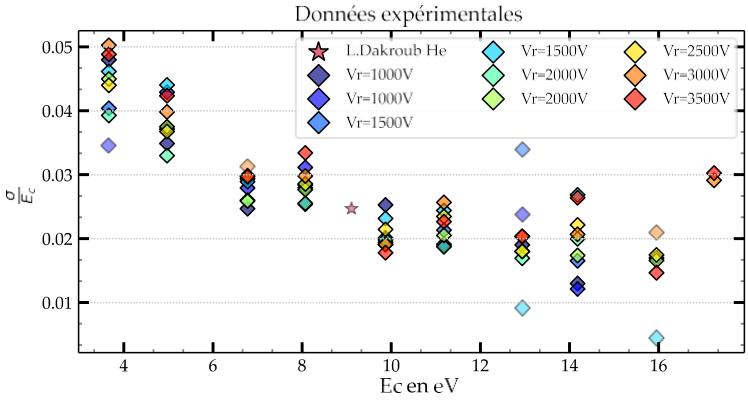


Figure 1: Resolution (obtained with standard deviation) of the 11-electrode VMIS as a function of the kinetic energy ofphotoelectrons

In molecules the presence of multiple energy bands makes the photoelectron spectra produced with high harmonic probe beams difficult to interpret. Therefore, a quasi-monochromatic XUV source would be beneficial for the interpretation of experiments. For this reason, high harmonic generation is driven by the second harmonic of the fundamental beam, at 400 nm. This allows the isolation of H3 around 9.3 eV. This photon energy will be used to probe the ultrafast dynamics of acetylacetone (2,4-pentandione) excited in the S2 state by a 270 nm pulse in a time-resolved experiment.

References:

[1] N. G. Kling *et al.*, « Thick-lens velocity-map imaging spectrometer with high resolution for high-energy charged particles », *J. Instrum.*, vol. 9, n° 05, p. P05005, mai 2014, doi: 10.1088/1748-0221/9/05/P05005.
[2] L. Dakroub *et al.* "Laser-dressed photoionization for the temporal characterization of attosecond pulses generated from plasma mirrors." *Eur. Phys. J. Spec. Top.* 232, 2055—2067 (2023). https://doi.org/10.1140/epjs/s11734-023-00817-5

23

Ultrafast spins and charges dynamics of the van der Waals antiferromagnet $FePS_3$

J. Besbas, V. Halte, J.L.P Wolff, A. Gloppe and M. Barthelemy

IPCMS-UMR7504 CNRS-Université de Strasbourg, 23 rue du Loess 67034 Strasbourg, France
barthelemy@unistra.fr

Abstract: An ambitious objective lies in achieving optical control of magnetic anisotropy in 2D magnets within ultrashort time scales. Toward that goal, we conduct a comprehensive investigation of sub-picosecond magnetic order and charges dynamics in $FePS_3$ van der Waals antiferromagnet, while the magnetic phase transition is crossed at 113 K.

1. Introduction

Sub-nm scale magnetic memory devices with controllable magnetic properties at THz frequencies are required to improve data processing and spintronics technologies. In such context, phosphorus trichalcogenides XPS_3 where X is a 3d transition metal (Mn, Fe, …) constitute an interesting class of 2D semiconductors. In particular, $FePS_3$ is an antiferromagnet below 113 K. It is known to have strongly coupled spin and charge orders and provide flexibility in terms of control of magnetism down to the 2D limit [1]. Its individual layers form hexagonal ionic crystals comprising magnetic Fe^{2+} and thiophosphate $P_2S_6^{2-}$ ions, with antiferromagnetically coupled zigzag spin structure.

2. Experiment

In this work, we provide a systematic study of the ultrafast spin and charges dynamics of a $FePS_3$ multilayer sample (Fig 1.a) as a function of the density of excitation and temperature. Using time resolved linear dichroism measurements [2], we conduct a comprehensive investigation of the ultrafast magnetic order dynamics while the magnetic phase transition is crossed.

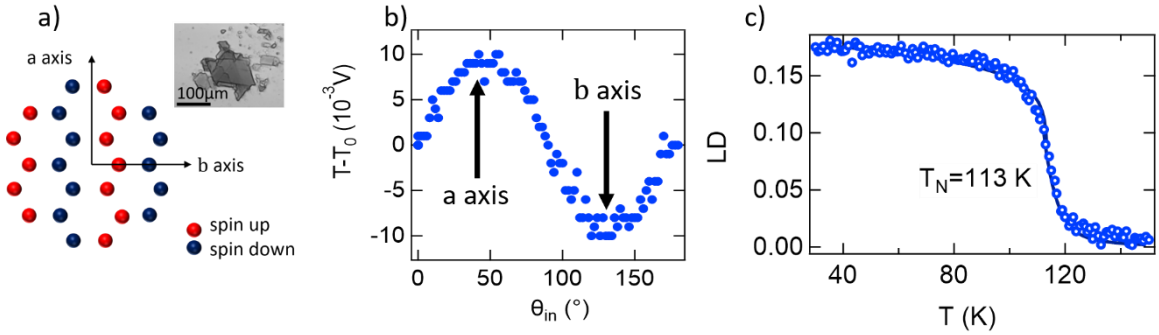


Figure 1: Principle of measurement. a) Spin structure of $FePS_3$. Inset: ~200 layers $FePS_3$ sample. b) Transmission as a function of probe linear polarization angle. c) static linear dichroism LD as a function of sample temperature.

The ultrashort excitation at 3 eV is resonant with optical transitions of thiophosphate ions, whereas the transmission dynamics, investigated at 1.5 eV, correspond to Fe^{2+} *d-d* transitions. As seen in Fig 1.b, by varying the linear polarization angle of the probe, the two dichroic axes of layers along zigzag (axis a) and armchair direction (axis b) of spin structure can be identified when the referenced transmission is maximal. The corresponding linear dichroism (LD) defined as difference of referenced transmission along axes a and b as a function of temperature is shown on Fig 1.c. The measurement is well reproduced by a 2D Ising model allowing to determine the Neel temperature as T_N=113 K (Fig 1).

3. Results and discussion

In the antiferromagnetic phase at 30 K, Fig 2.a shows a strong decreasing of transmission followed by a slow recovery. We will delve into the transfer process of hot electrons from the excited $P_2S_6^{2-}$ ions to the *d* bands of the Fe^{2+} ions. The corresponding magnetic order dynamics behaves very differently, with a three-step decay (Fig 2.b). The distinct contributions to demagnetization including crystalline field modification will be discussed in details. In particular, the corresponding transient modification of crystal field leads to a change of magnetic anisotropy via spin-orbit coupling [3]. Such control of magnetic state opens appealing prospects for controlling the magnetic anisotropy at THz frequencies.

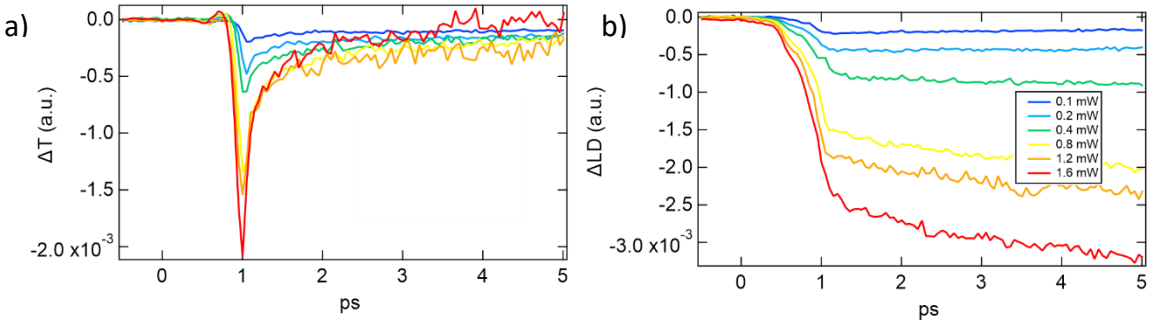


Figure 2: Pump-probe measurements in $FePS_3$ at 30 K vs pump fluence. a): Transient transmission (□T). Inset: ~200 layers thick $FePS_3$ flake. b) Transient linear dichroism (□LD) associated to the magnetic order dynamics.

References

[1] Gibertini, M. *et al.*, Nat. Nanotechnol. **14**, 408–419 (2019).

[2] Mertens, F *et al.*, Adv. Mater. **35**, 2208355 (2023).

[3] Matthiesen, M. *et al.,* Phys. Rev. Lett. **130**, 076702 (2023).

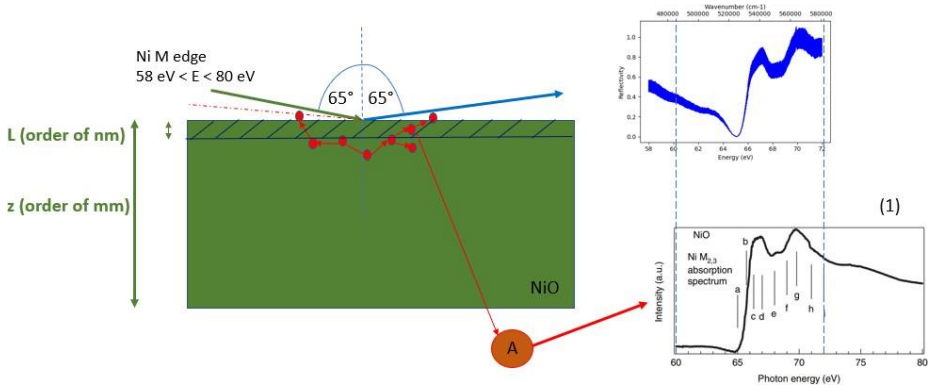
24

Obtaining optical constants from absorption spectroscopy and reflectivity data

Amina Alic[1], Romain Cazali[2], Romain Geneaux[2], Gheorghe Sorin Chiuzbaian[1]

1. Sorbonne Université, CNRS, Laboratoire de Chimie Physique-Matière et Rayonnement, 75005, Paris, France
2. Université Paris-Saclay, CEA, CNRS, LIDYL, 91191, Gif-Sur-Yvette, France

Optical constants, such as the dielectric function or refractive index describe how materials respond to incident electromagnetic waves. Obtaining these constants from experimental measurements is crucial for data analysis and physical interpretation of results, forming the foundation for analysis of time-resolved data. In this abstract we will present computational methods used for extracting optical constants from soft x-ray reflectivity data and total electron yield (TEY) x-ray absorption spectroscopy (XAS) data for bulk samples, using NiO at Ni M-edges as a specific example. Reflectivity spectra were acquired at ATTOLAB on the FAB1bis beamline, which provides extreme ultraviolet light through high harmonic generation of 800 nm, 5 fs laser pulses. The TEY XAS data were acquired at the Swiss Light Source using synchrotron radiation, as described in the literature [1]. In order to extract optical constants from reflectivity data, we can employ simple modelling with Lorentz oscillators. For better precision, we can apply a variational dielectric function "correction" on top of the Lorentz model [2]. In order to ensure an accurate Kramers-Kronig transformation, we have also incorporated the tabulated Henke data outside of the resonant energy region. Regarding TEY XAS data, we need to account for several aspects, such as efficiency of creating secondary electrons and surface effects in order to create a viable model [3] that we can then use to iteratively compare to the reflectivity data (via Kramers Kronig transformation [4]). We will show the limitations and practical difficulties of this approach.



Obtaining reflectivity and TEY XAS data from a bulk sample

References:

[1] Chiuzbăian, S. G., Ghiringhelli, G., Dallera, C., Grioni, M., Amann, P., Wang, X., Braicovich, L. & Patthey, L. (2005). Localized electronic excitations in NiO studied with resonant inelastic x-ray scattering at the Ni M threshold: Evidence of spinflip. *Physical Review Letters*, *95*(19), 197402.

[2] Kuzmenko, A. B. (2005). Kramers–Kronig constrained variational analysis of optical spectra. *Review of ScientificInstruments*, *76*(8).

[3] Jansing, C., Wahab, H., Timmers, H., Gaupp, A., & Mertins, H. C. (2018). Soft X-ray refractive index by reconciling total electron yield with specular reflection: experimental determination of the optical constants of graphite. *Journal of SynchrotronRadiation*, *25*(5), 1433-1443.

[4] Watts, B. (2014). Calculation of the Kramers-Kronig transform of X-ray spectra by a piecewise Laurent polynomialmethod, *Optics Express*, *22*(19), 23628-23639.

25

Holographic storage of ultrafast photonic qubit in molecules

A. Voisine, F. Billard, O. Faucher, P. Béjot, and E. Hertz

Laboratoire Interdisciplinaire Carnot de Bourgogne (ICB) Team : Processus Femtosecondes & Lasers intenses (PFL)

We demonstrate that ultrashort spatially structured beams can sculpt a sample of gas-phase molecules like a 4D material so as to produce a spatial pattern of aligned molecules whose shape and temporal evolution allow to restore the spatial light information on a time-delayed reading pulse. To do so, the spatial phase and amplitude information of ultrashort light beams is encoded into rotational coherences of molecules by exploiting the interplay between spin angular momentum and orbital angular momentum. The field-free molecular alignment resulting from the interaction leads to an inhomogeneous spatial structuring of the sample allowing to re-embed the encoded information into a time-delayed probe beam. The demonstration is conducted in CO_2 molecules. Besides applications in terms of THz bandwidth buffer memory, the strategy features interesting prospects for establishing versatile opticalprocessing of OAM fields, for studying various molecular process or for designing new photonic devices enabling to impart superpositions of OAM modes to light beams.

26

Few-cycle optical vortices for strong field physics

Matthieu Guer[1], Martin Luttmann[1], Jean-François Hergott[1], Fabien Lepetit, Olivier Tcherbakoff[1], Thierry Ruchon[1], Romain Géneaux[1,2]

1. Université Paris-Saclay, CEA, LIDYL, 91191 Gif-sur-Yvette, France
2. CY Cergy Paris Université, CEA, LIDYL, 91191, Gif-sur-Yvette, France

Laser beams carrying orbital angular momentum (OAM), such as optical vortices (OVs), have becomeubiquitous in many areas of photonics [1]. In strong-field laser-matter interaction, OVs are now used togenerate novel types of radiation, such as attosecond twisted beams in the extreme ultraviolet (XUV) orrelativistic electron vortex beams. However, one type of light beam has yet to be produced: OVs with few- cycle pulse duration and enough energy to reach the strong-field regime.

Few-cycle vortices have been previously produced at low intensity, by shaping an already post-compressed pulse. The drawback is the need of a broadband device, to transfer OAM to a wide spectrum. In our study[2], we demonstrate another method, which is to start from a regular Ti:sapph Gaussian beam at 25fs, onwhich it is easier to imprint OAM. For this, we use a spiral phase plate designed at 800nm. We then usedan argon-filled stretched hollow core fiber to broaden its spectrum and finally compress the pulse. This has the advantage of using common instruments, and being easily scalable in energy.

We achieved a pulse duration of 5.5fs (2.1 optical cycles), measured by dispersion scan. Furthermore, we demonstrate that the vortex charge is transferred to the broadened spectrum with a method based on spatially resolved Fourier-transform spectroscopy. The energy transmission is 25%, resulting in a 0.5mJoutput, which needs to be improved. Nonetheless, given our short pulse duration, we can reach high irradiance values ($1.5x10^{14}W/cm^2$ with a 1m focal lens), enough to drive strong nonlinear processes. Forinstance, using this type of beam in high-harmonics generation could give rise to isolated attosecond vortices, which to our knowledge have not been described yet, and would be useful for probing ultrafastdynamics of chiral phenomena.

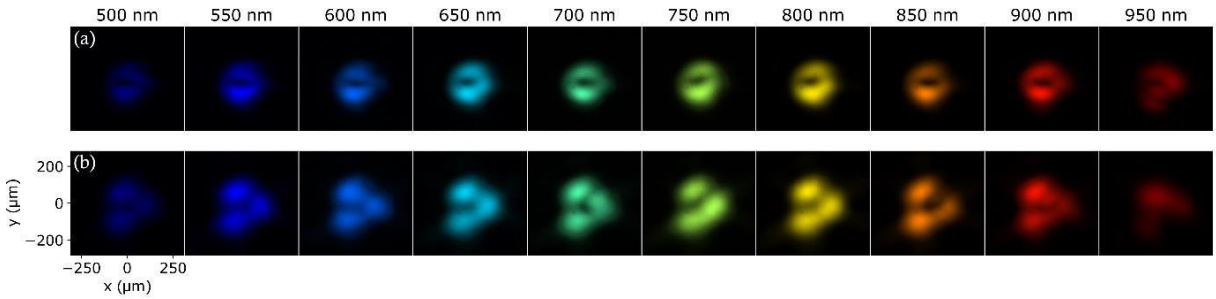


Figure 1: (a) Focus of the beam and (b) diffraction pattern by a triangular slit for several wavelengths, retrieved by Fourier-transform spectroscopy. The three-dot diffraction pattern is characteristic of a beam whose orbital angular momentum is dominated by $P = \pm 1$.

References:

[1] Dennis, M. R., O'Holleran, K. & Padgett, M. J. Singular optics: optical vortices and polarization singularities. Prog. Opt. **53**, 293–363 (2009).

[2] M. Guer et al. "Few cycle vortices for strong field physics", Opt. Lett. https://doi.org/10.1364/OL.509802 (2023)

27

Ultrafast photoreaction C=C double bond Isomerization dynamics of molecular switches

Matthew Mgbukwu [a], Valerio Giuso [a], Stefano Crespi [b][c], Matteo Mauro[a], Stefan Haacke[a], Jérémie Léonard*[a]

[a] Institut de Physique et Chimie des Matériaux de Strasbourg, Université de Strasbourg, CNRS UMR 7504, Strasbourg, France

[b] Stratingh Institute for Chemistry, Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands

[c] Department of Chemistry, Ångström Laboratory, Uppsala University, Box 523, 751 20 Uppsala, Sweden

jeremie.leonard@ipcms.unistra.fr

Bistable molecular switches play a crucial role in the photocontrol of molecular motion and function in a range of applications such as photopharmacology, molecular electronics, and molecular machines. Efficient molecular switches have a high photoisomerization quantum yield (QY). To explore the molecular design rules which control this QY, we introduce two parallel strategies, (a) Chemical modification of the molecular switch, by introducing an electron-donating moiety on an oxindole switch (see figure1A), which creates an electronic push-pull effect. (b) We further explore the influence of the environment (solvent and macrocyclic cage) on the biomimetic indanylidene pyrroline (IP) Schiff base [2], [3] inspired by the retinal chromophore in rhodopsin protein[4]. We employ time- resolved UV-Vis transient absorption spectroscopy to observe the C=C photoisomerization dynamics of both molecular switches in various solvents. For the oxindole switch, the push-pull effect, already proposed be responsible for the large QY of 50%[1], is futher enhanced by the deprotonation of the phenol moiety. While we observe a significantly shorter excited state lifetime, the QY of phenolate oxindole derivative remains to be quantified (on going wok). For the neutral IP compound encapsulated in the macrocylic cage, the photoreaction dynamics appears very similar as compared to the protonated IP in water but the QY is enhanced (preliminary results). These findings will deepen our comprehension of the influence of the molecular design rules on the QY of photoswitches and provide valuable insights for tailored applications in photopharmacology and molecular motors.

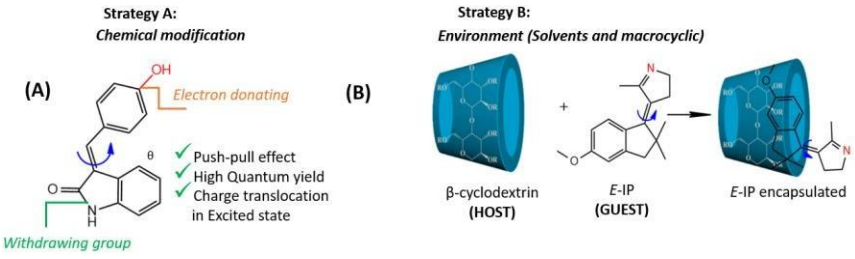


Figure1: two strategies to reveal how to control/optimze the photoisomeriztion mechanism and QY: A. Oxindole switch: tuning electronic structure (push-pull effect) B. IP biomimetic compound: tuning the interaction with the envirionement.

References:

[1] D. Doellerer, D. R. Pooler, A. Guinart, S. Crespi, and B. L. Feringa, "Highly Efficient Oxindole-Based Molecular Photoswitches," *Chem. Eur. J.*, p. e202301634, 2023.

[2] M. Gueye, M. Paolino, E. Gindensperger, S. Haacke, M. Olivucci, and J. Léonard, "Vibrational coherence and quantum yield of retinal- chromophore-inspired molecular switches," *Faraday Discuss.*, vol. 221, pp. 299–321, 2020.

[3] M. Gueye *et al.*, "Engineering the vibrational coherence of vision into a synthetic molecular device," *Nat. Commun.*, vol. 9, no. 1, p. 313, 2018.

[4] G. Schneider, R. Diller, and M. Stockburger, "Photochemical quantum yield of bacteriorhodopsin from resonance Raman scattering as a probe for photolysis," *Chem. Phys.*, vol. 131, no. 1, pp. 17–29, 1989.

[5] C. R. Reddy, V. Ganesh, and A. K. Singh, "E–Z isomerization of 3-benzylidene-indolin-2-ones using a microfluidic photo-reactor," *RSC Adv.*, vol. 10, no. 48, pp. 28630–28634, 2020.

28

Femtosecond laser induced dynamics of protonated reserpine

E. Prost[1], S. Palakkal[1], Y. Hu[1], A. Boyer[1], M. Hervé[1], R. Bredy[1], I. Compagnon[1], F. Lépine[1]

1. Institut lumière Matière, UMR 5306 Université Lyon 1 – CNRS, Université de Lyon, 69622 Villeurbanne, France

Ultra-violet excitation of molecules generally involves the excitation of a chromophore carrying a localizedelectronic excited state. The relaxation of such an excited state is a complex process that can involve ultra- fast charge transfer, structural rearrangement and energy dissipation between the electronic and/or the nuclear degrees of freedom. Such mechanisms can be studied through ultrafast pump-probe experiments in the gas-phase.

We explore photo-physical properties of protonated reserpine by coupling an electrospray ionization source (ESI) and mass spectroscopy (MS) with a UV-IR ultrafast pump-probe experiment [1]. With this setup, we perform "on-the-fly" experiments ensuring that molecules interact with one single laser pulse.

In UV-IR ultrafast pump-probe experiments we observed non-adiabatic relaxation of the UV excited protonated reserpine through conical intersection in about 4 ps [2]. This shows that the reserpine molecule efficiently converts UV excitation and electronic energy into internal vibrational heating. This result could explain the fluorescence quenching observed in the molecule [3].

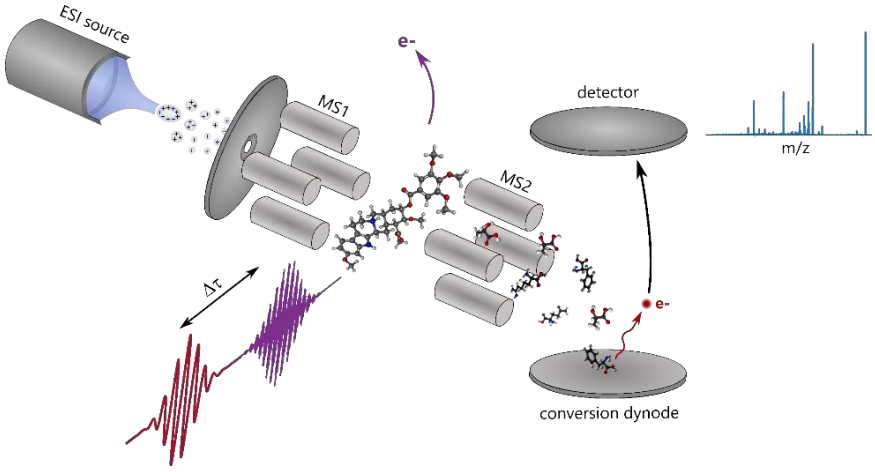


Figure : Schematic diagram of an "on-the-fly" UV-IR pump-probe mass spectroscopy experiment

References:

[1] Hervé M., *et al* 2019 *J. Phy. Chem. Lett.* **10** 2300

[2] Hervé M., *et al* 2022 *Adv. Phys.-X* **7** 212328

[3] Savory B and Turnbull 1983 *J. Photochem.* **23** 171-181

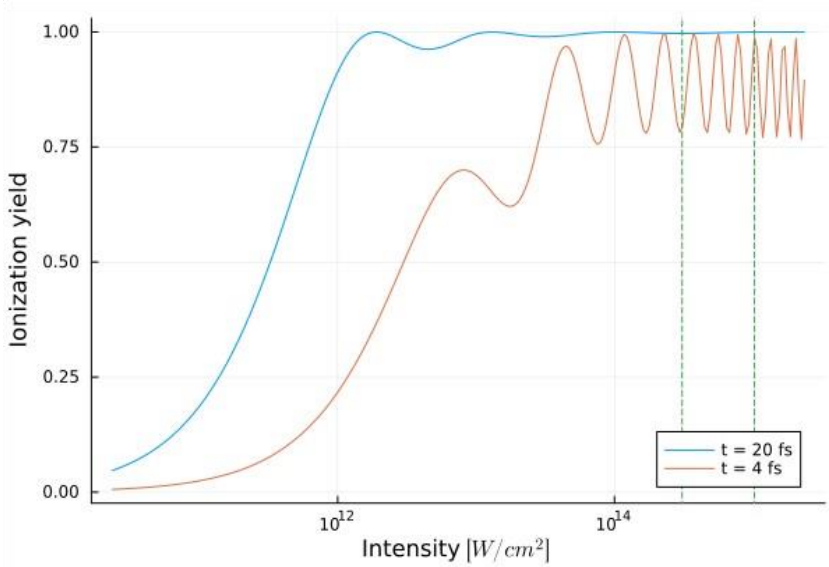
29

Theoretical Study of the Resonant Auger decay in diatomic molecules

K.Marmorat[1], M.Labeye[1], …

1. PASTEUR, Département de chimie, École normale supérieure, PSL University, Sorbonne Université, CNRS, 75005 Paris, France

With the development of new instruments, the resonant auger decay has been studied with high definition and in intense field. In this work, we propose a full time dependent theory of the process, whose equation of motion can be integrated numerically, allowing us to give insight on the ultrafast dynamics of the highly excited molecules as well as allowing us to do simulation in the non-linear regime. Reaction of the molecules with ultrafast pulses, on the same order of magnitude than the excited state lifetime, can lead to oscillations in the total ionization. Also, the effect of nuclear motion, known as lifetime broadening, is shown to be due to the coupling of the potential energy surfaces.



Ionization yield following the resonant auger decay of N_2 molecules excited by a Gaussian pulse of frequency accorded to the first vibrational level of the excited state, of lifetime Γ= 4.8fs. One can see that oscillations in the Ionization yield appears for sufficiently short impulsion, when the Rabi coupling between the excited and ground state become important.

References:

(1) Demekhin, P. et. al. Phys. Rev. A 2011, 84 (3), 033417
(2) Gortel, Z. W. et al. Phys. Rev. A 1998, 58 (2), 1225–1246

30

TR-ARPES under uniaxial pressure to demystify the relation between quantum criticality and van-Hove singularity in hole-doped cuprates

B. Michon[1], S. Benhabib[1], P. Senzier[1], C. Pasquier[1] and M. Marsi[1].

[1] *Laboratoire de Physique des Solides - CNRS, Université Paris-Saclay, Orsay, France*

The origin of the superconducting pairing mechanism in cuprates has remained an enigma since their discovery in 1986. New experimental breakthroughs have recently revealed important signatures suggesting the presence of quantum criticality in the phase diagram of hole doped cuprates, where the pseudogap state vanishes in doping at the critical point p* and zero temperature:

1) Linear-in-temperature resistivity $\rho \propto T$ [1]
2) Strong effective mass m* enhancement and an unusual logarithmic divergence in temperature at p* for the normal state specific heat [2][3]
3) Linear scaling law for the optical scattering rate $\tau(\omega,T)$ in energy ω and temperature T and $\log(T)$ dependence for the effective mass ratio $m^*/m(\omega \to 0,T)$ in infrared spectroscopy [4].

Nevertheless, a Lifshitz transition in hole doping modifying the Fermi surface topology from hole-like to electron-like also occurs around the same doping p* associated to a saddle point in the band structure crossing the Fermi level [5][6]. This saddle point, called a van Hove singularity, gives rise to a peak in the density of states, a $\log(T)$ dependence for the effective mass and a linear-in-temperature resistivity exactly like quantum criticality. The presence of a van Hove singularity thus casts doubts on the real presence of quantum criticality.

To elucidate the relations between van-Hove singularity and quantum criticality in hole doped cuprates, we propose to develop an experimental combination of static and time-resolved ARPES (TR-ARPES) techniques with a uniaxial pressure sample environment. The static ARPES technique is able to detect the location of the Lifshitz transition in doping causing the van Hove singularity. With the TR-ARPES technique at femtosecond scale, we can detect the relaxation rate of out-of-equilibrium electronic excitations. At a quantum critical point, this relaxation rate should satisfy a particular scaling law in temperature. We are thus sensitive to the position of the quantum critical point. By the application of uniaxial pressure, being able to push van-Hove singularities through the Fermi level [7], we could discriminate if all the aforementioned peculiar signatures are associated to band structure Physics – van-Hove singularity – or in fact to quantum criticality, and assess how these two physical phenomena are linked and could give birth to superconductivity.

References

[1] R.A. Cooper, Y. Wang, B. Vignolle *et al.*, Science **323**, 603 (2009).

[2] B. Michon, C. Girod, S. Badoux *et al.*, Nature **567**, 218 (2019).

[3] C. Girod, D. LeBœuf, A. Demuer *et al.*, Phys. Rev. B **103**, 214506 (2021).

[4] B. Michon, C. Berthod, C.W. Rischau *et al.*, Nat. Comm. **14**, 3033 (2023).

[5] Y. Zhong et al., PNAS 119, 32 (2022).

[6] D.C. Peets et al., New Jour. of Phys. **9**, 2 (2007).

[7] V. Sunko et al., Nat. Quant. Mat. **4**, 46 (2019).

31

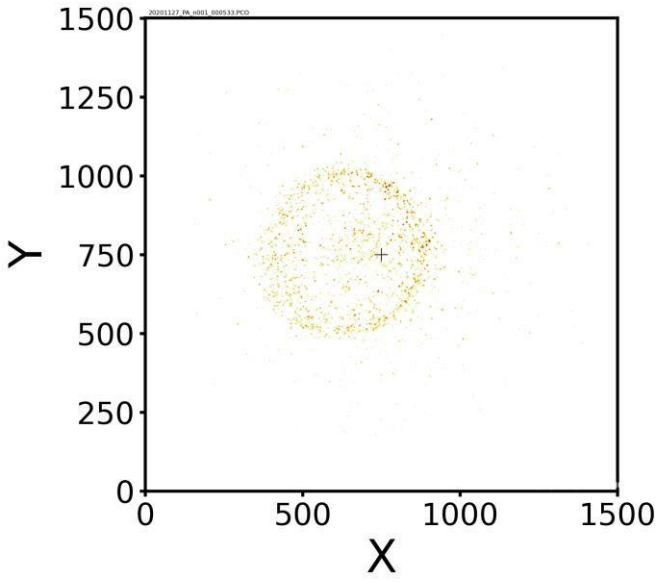
Photoionization dynamics of isolated tryptophane nanoparticles

G. Gallician[1], A. Quentin-Derlich[2], M. Dalui[1], A. Lietard[1], B. Cunha de Miranda[1], S. Habka[1], A. Klisnick[2], L. Barreau[2], L. Poisson[1,2]

1. LIDYL, CEA, CNRS, Université Paris Saclay, CEA Saclay 91191 Gif-sur-Yvette, France
2. ISMO, Université Paris-Saclay, Faculté des Sciences d'Orsay, CNRS, F-91405 Orsay, France

Isolated nanoparticles are little volumes of condensed mater that are introduced into vacuum. Because of the high density, they are several order of magnitude more sensitive to a laser pulse than isolated molecules. In the framework of gas phase studies, the interaction of an ultrashort femtosecond laser pulse with nanoparticles leads to a high ionization rate. Then, the freed electrons oscillate with the electric field and give rise to collisions with the constituents of the particles. This enhances the number of ions and electrons until formation of a plasma at the nanoscale[1].

Several parameters have been identified to play a role the nanoplasma dynamics, such as size, shape and refractive index of nanoparticles[2,3]. Our approach consists in using a Velocity Map Imaging spectrometer (VMI) to study the anisotropy of ejection of the electrons and ions from this nanoplasma as the function of the size. Here, we will focus on the ejection of the H- anion in the case of tryptophane nanoparticles irradiated with 266, 400 and 800 nm laser pulses around 10^{14} W.cm^{-2}.



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

References:

(1) Fennel, T.; Meiwes-Broer, K. H.; Tiggesbäumker, J.; Reinhard, P. G.; Dinh, P. M.; Suraud, E. *Reviews of Modern Physics* **2010**, *82*, 1793.

(2) Antonsson, E.; Gerke, F.; Merkel, L.; Halfpap, I.; Langer, B.; Rühl, E. *Phys. Chem. Chem. Phys.* **2019**, *21*, 12130.

(3) Ban, L.; Yoder, B. L.; Signorell, R. *Annu. Rev. Phys. Chem.* **2020**, *71*, 315.