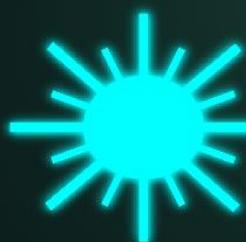




Laserlab User Meeting

6-8 September 2015, Coimbra, Portugal



Illuminate- Lasers in the Year of the Light

Book of Abstracts



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LASERLAB-EUROPE is the consortium of the major European laser research organisations. In 2015, LASERLAB-EUROPE will complete its third European programme and will enter a new phase of collaboration until 2019. In a very competitive call the consortium has been successful in securing EC funding of 10 million euros in Horizon 2020.

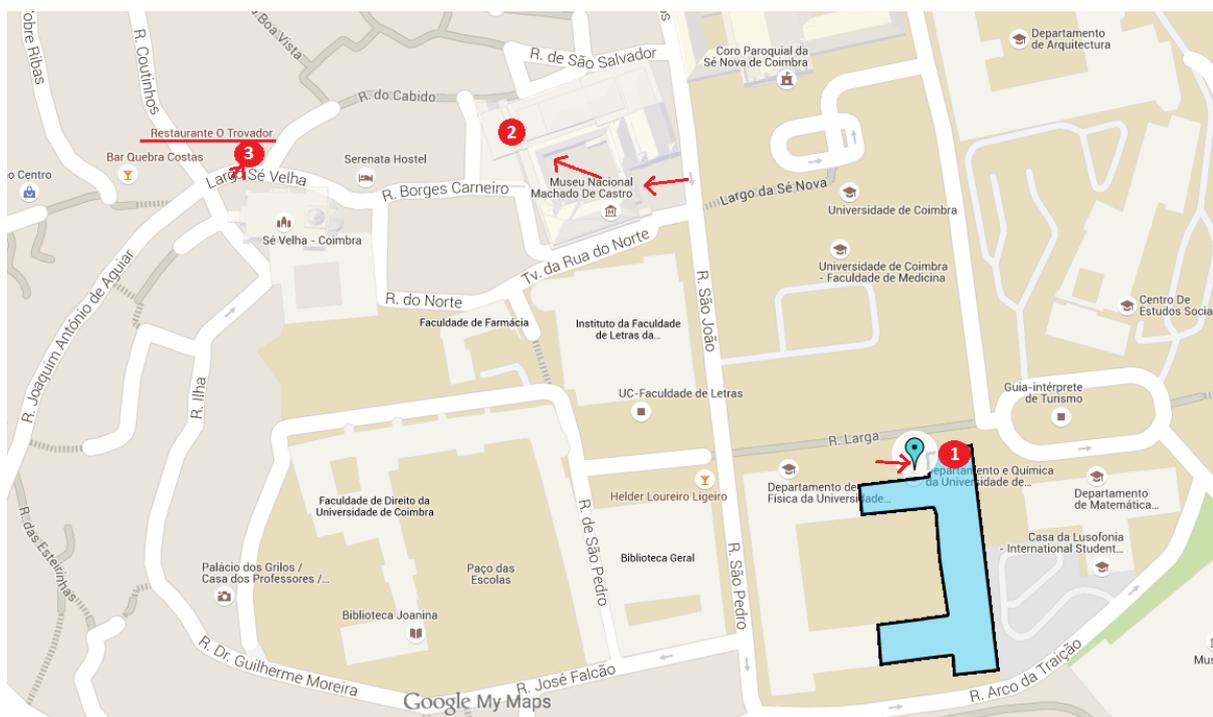
In the upcoming 4-year phase, starting in December 2015, LASERLAB-EUROPE will comprise 33 of the leading European laser infrastructures and, together with subcontractors and associate partners, involve coordinated activities in 21 countries. The members offer free access to key complementary laser facilities in Europe with performances at the international forefront of laser technology. This allows guest scientists from academia as well as from industry to carry out leading-edge research for the advancement of knowledge in a wide range of scientific domains, thus serving a very broad and interdisciplinary community.

The User Meeting entitled “**Illuminate – Lasers in the Year of Light**” takes place at the transition between the third and fourth LASERLAB European programs and coincides with the **International Year of Light and Light-based Technologies (IYL)**. In proclaiming IYL, the UN has recognized the importance of raising global awareness about how light-based technologies promote sustainable development and provide solutions to global challenges in energy, education, agriculture and health. Light plays a vital role in our daily lives and is an imperative cross-cutting discipline of science in the 21st century. Lasers are the ultimate example of mastering light for the benefit of our societies.

The 47 registered participants from 15 countries will present 22 talks covering subjects related to biophotonics, structural dynamics, advanced laser sources and ultrafast probing. This cross-section of the thousands of users that have benefited from access to leading European laser infrastructures for over a decade, illustrates the success of transnational access in a European distributed infrastructure, pioneered by LASERLAB-EUROPE.

Hosting this User Meeting at the Coimbra LaserLab was possible thanks to the efforts of many individuals. The labs offering access were particularly effective in promoting this meeting in their users community and the users responded promptly. The organizers are very grateful to Didier Normand and Daniela Stozno for their advice and responsiveness. A very special thanks is due to Kamila Mentel for her enthusiasm and competence in the organization of the meeting.

Luis Arnaut
Host of the User Meeting
Coimbra LaserLab, Portugal



1. Chemistry Department

University of Coimbra
Rua Larga
Coimbra



2. Loggia restaurant

in the National Museum
Machado de Castro
Largo Dr José Rodrigues
Coimbra



3. O Trovador restaurant

Largo Sé Velha nº 15-17
Coimbra

Scientific programme sessions

The scientific programme sessions will take place in the Seminar Room E-35 (second floor) in the Chemistry Department.

Access Board Meeting

Access Board Meeting will take place in the Room C-18A (ground floor) in the Chemistry Department. The volunteers will assist participants finding their way indicating the entrance to the room.

Social Programme

Guided visit to the University (Sunday, 6 September)

Laserlab User Meeting will start with guided tour to the Palace of Schools of the University of Coimbra classified as UNESCO World Heritage.

Meeting point: entrance of Chemistry Department at 16:50 on 6 September.

Welcome reception (Sunday, 6 September)

Participants have opportunity to meet in friendly atmosphere during Meeting Welcome Reception, which begins at 19:00, in the **Loggia** (Restaurant in the National Museum Machado de Castro).

Lunch

Lunches will be served in **Loggia**, between 12:30 and 14:00 on 7 and 8 September.

Dinner

Dinner will take place in restaurant **O Trovador**, located in Old Town, a couple of minutes walk from Meeting Venue. Dinner starts at 20:00 on 7 September (Monday) and will be accompanied by Fado- Portuguese music.

Organising committee:

Arnaut Luis

Mentel Kamila

Stozno Daniela

Sunday, September 6th

16:00-17:00 Registration

17:00-18:30 Guided visit to the historical part of the university

19:00 Welcome reception

Monday, September 7th

8:30-12:00 Registration

9:15-9:40 Opening addresses

9:40-10:10 **Didier Normand**- Chair of The Access Board

10:10-10:40 Coffee break

Session I: Biophotonics and health applications of lasers

Chair: Luis Arnaut (Portugal)

10:40-11:00 **Janusz Dąbrowski**: Photodynamic Therapy (PDT): basic mechanisms, clinical outcomes, and future research direction / CLL

11:00-11:20 **Albena Daskalova**: Immobilized biopolymer thin films modified by temporally shaped femtosecond pulses for cell guidance improvement / ULF-FORTH

11:20-11:40 **Domenico Doria**: Radiobiology with laser-accelerated ion bursts / LULI

11:40-12:00 **Zbignevs Marcinkevics**: Blood perfusion changes in exercising and non-exercising muscle during unilateral static exercise using diffuse correlation spectroscopy technique (DCS) / ICFO

12:00-12:20 **Judy Zouaoui**: Toward In Vivo and In Situ time domain reflectance Diffuse Optical Tomography thanks to time gated SPAD systems / CUSBO

12:20-14:00 Lunch

Session II: Structural dynamics and materials

Chair: Rui Fausto (Portugal)

14:00-14:20 **John Bartholomew**: High spectral resolution spectroscopy of individual rare-earth-ion doped nanoparticles / LLC

14:20-14:40 **Stefano Colonna**: Ultrafast photoemission spectroscopy study of the electron-lattice coupling in the 2D metallic system alpha-Sn/Ge(111) / CELIA

14:40-15:00 **Javier Solis/ Mario Garcia-Lechuga**: Simultaneous time-and-space resolved reflectivity and interferometric measurements of dielectrics excited with femtosecond laser pulses / SLIC

15:00-15:20 **Junqing Shi**: Solid State Luminescence Enhancement in Dicyano-Distyrylbenzenes: Intra- and Intermolecular Contributions / CLL

15:20-15:40 **Kestutis Staliunas**: Photonic crystal spatial filters / VULRC

15:40-16:00 **Igor Reva**: Near-Infrared Laser-Induced Generation of Higher-Energy Conformers in Matrix Isolated Compounds / CLL

14:00-16:00

Access Board Meeting

16:00-16:40

Coffee break

Session III: Advanced laser sources
Chair: Oldrich Renner (Czech Republic)

16:40-17:00 **Andrzej Bartnik:** Photoionized plasmas induced by intense soft X-ray and extreme ultraviolet pulses / PALS

17:00-17:20 **Brendan Dromey:** Polarisation gating of high harmonic generation from relativistic laser plasmas / IOQ

17:20-17:40 **Vaclav Kubecek:** 39 mJ Energy, Picosecond Ho:YLF Cryogenic Chirped Pulse Amplifier for Mid-Infrared OPCPA / ICFO

17:40-18:00 **Florian Mollica:** Efficient laser production of energetic neutral beam / MBI

18:00-18:25

LaserLab Portugal Roadmap

18:25-18:55

Visit to the Coimbra LaserLab facility

20:00

Gala dinner

Tuesday, September 8th

Session IV: Ultrafast probing and high energy lasers I
Chair: Istvan Földes (Hungary)

9:30-9:50 **Amelle Zair:** Temporal flat top synthesis for high order harmonic generation / CELIA

9:50-10:10 **Barry Bruner:** Two-dimensional phase resolved high harmonic spectroscopy of molecular vibrations / CELIA

10:10-10:30 **Livia Lancia:** A plasma based laser amplifier / LULI

10:30-11:00

Coffee break

11:00-12:20

Round table

Chair: Jouko Korppi-Tommola (Finland)

12:20-14:00

Lunch

Session V: Ultrafast probing and high energy lasers II
Chair: Rosa Weigand (Spain)

14:00-14:20 **Franck Lepine:** Short XUV pulses for ultrafast molecular physics and astrochemistry / MBI

14:20-14:40 **Anja Roeder:** Femtosecond dynamics of isolated radicals in the gas phase / SLIC

14:40-15:00 **Tatjana Stoll:** Ultrafast vibrations of small atomically defined thiolate gold clusters studied by femtosecond transient absorption spectroscopy / CUSBO

15:00-15:20 **Jan Vanda:** Laser induced damage threshold measurements of optical components for high-energy HiLASE lasers / VULRC

Closing remarks, end of meeting

15:30-16:00

Coffee break

THE LASERLAB-EUROPE ACCESS PROGRAM

Didier Normand¹, Access Board chairperson

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The LASERLAB-EUROPE Transnational Access programme aims at providing European scientists access to world-class laser research facilities all over Europe:

- to carry out research in a large variety of research domains,
- free of charge, including travel and accommodation.
- on the basis of scientific excellence of a research proposal.

An overview of the LASERLAB-3 access programme (June 2012- November 2015) will be presented. This programme is currently very successful, with all contractual commitments being widely fulfilled already 3 months before the end of the contract.

The LASERLAB-4 contract will start on December 1st, 2015 and will last 4 years. The main evolutions of the access programme will be presented, with a highlight on the new access providers.

PHOTODYNAMIC THERAPY (PDT): BASIC MECHANISMS, CLINICAL OUTCOMES, AND FUTURE RESEARCH DIRECTION

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²Chemistry Department, University of Coimbra, Rua Larga, Coimbra, Portugal

Photodynamic therapy (PDT) is a clinically approved, minimally invasive therapeutic procedure, which is entering the mainstream of the cancer treatments. It requires a proper laser, a photosensitizer and oxygen to trigger photochemical and molecular mechanisms that lead to the primary tumour destruction and provide long-lasting protection against metastasis. In this work we show how the spectroscopy, photophysics, photochemistry and pharmacokinetics of photosensitizers determine mechanism of cell death and clinical protocols. Special attention is given to the description of the properties of NIR absorbing photosensitizers, because they provide the grounds to understand the molecular mechanisms and photodynamic efficacy.

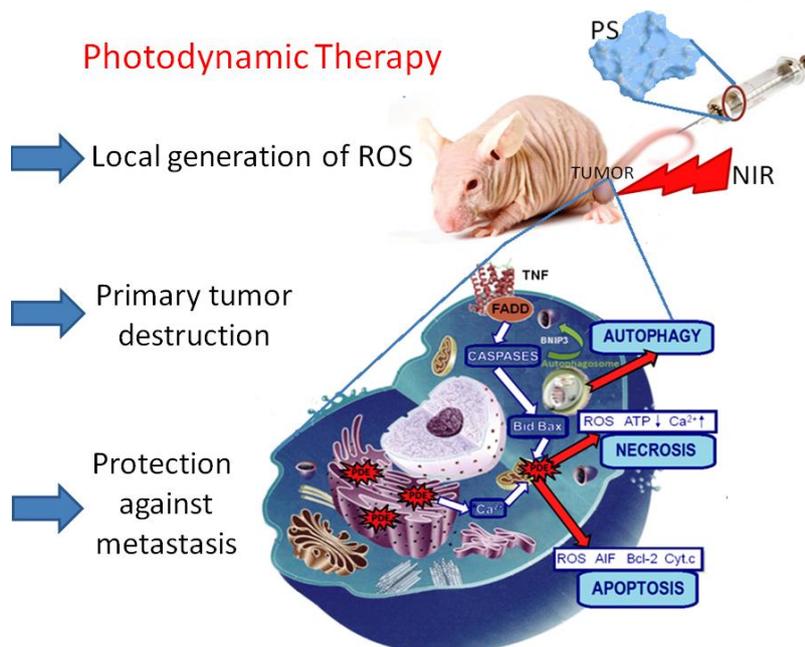


Figure 1: Schematic illustration of Photodynamic Therapy (PDT) of Cancer.

PDT with bacteriochlorins offers the advantage of the unique targeting offered by laser light and optical fibres to direct the strongest effect to the primary tumours and yet the ability to potentiate the systemic effects of chemotherapies and immunotherapies.

Acknowledgment: NCN and MNiSW are acknowledged for grants 2013/11/D/ST5/02995 and IP2014008573.

[1] Dąbrowski J. M.; Arnaut, L. G. *Photochem. Photobiol. Sci.* **2015**, *14*, in press.

[2] Rocha L. B.; Gomes-da-Silva L.; Dąbrowski J. M.; Arnaut, L. G. *Eur. J. Cancer* **2015**, *51*, 1822–1830.

IMMOBILIZED BIOPOLYMER THIN FILMS MODIFIED BY TEMPORALLY SHAPED FEMTOSECOND PULSES FOR CELL GUIDANCE IMPROVEMENT

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Cell–biomaterial interaction is a complex mechanism comprising interplay of different processes expressed in regulation mechanisms of cell’s adhesion, migration, proliferation, growth and differentiation, cell responses to surface morphology, knowledge of biochemical and biomechanical properties of the biomimetic material. Material’s surface topography is one of the most important aspects to construct a novel template for tissue regeneration. The scaffold constructs are designed to improve cell guidance and cell proliferation and for the support of tissue development. The efforts in this field are directed towards advancements in the fabrication of highly functional biomaterials. Employing femtosecond laser modification method for application in tissue engineering opens up possibility to control the surface topography. Pulse shaping technique was employed for ultra-short laser irradiation of biopolymer thin films. This method permits a gradual energy delivery into the sample due to the possibility of temporal redistribution of applied energy in the laser pulse. The influence of different pulse shapes and delay times between consecutive pulses was examined with regard to evolution of surface morphology via field emission scanning electron microscopy (FESEM). Femtosecond pulses–biopolymer interaction initiate bio-functionalization, by tuning the surface properties to creation of various topography features of biopolymers. In this study was shown that initial cell-interface interaction reveal a preferable cell migration and significant increase in cell orientation on fs laser-modified surface array Fig.1.

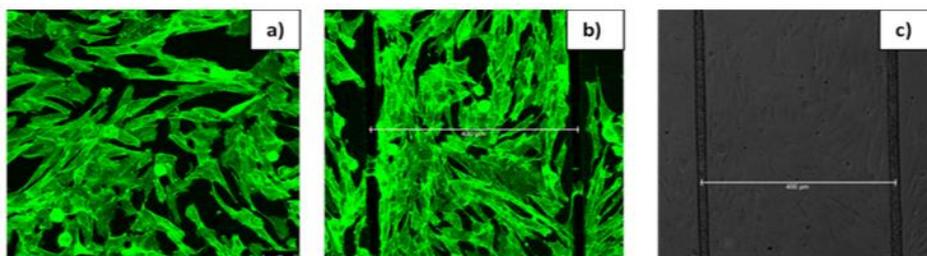


Figure 1: Confocal laser microscopy pictures of MG63 cells cultured for 3 days on the non-irradiated (a) and laser processed (b, c) collagen/elastin thin film surfaces.

It was demonstrated that surface topography has an important effect on cells mobility and that cells are able to reorganize themselves in relation to surface nanofeatures.

Acknowledgements: This work was supported by Integrated Laserlab Europe Initiative FP7 of EU through project № ULF-FORTH002013/2014 and COST action MP1301, NEWGEN.

[1] Ortiz, R.; Morreno-Flores, S.; Quintana, I.; Vivanco, M.; Sarasua, J. R.; Toca-Herrera, J. L. *Mat. Sci. and Eng. C* **2014**, *37*, 241-250.

RADIOBIOLOGY WITH LASER-ACCELERATED ION BURSTS

Domenico Doria¹, Deborah Gwynne¹, Giacomo Candiano², Pankaj Chaudhary³, G.A.P.Cirrone², Renata Leanza², Lorenzo Manti^{4,5}, Francesca Perozziello^{4,5}, Kevin Prise³, Lorenzo Romagnani⁶, Francesco Romano², Antonella Tramontana², Marco Borghesi¹

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⁶LULI, École Polytechnique, CNRS, CEA, UPMC, 91128 Palaiseau, France

Hadron-therapy is successfully used worldwide in treating localised tumors, although the cost of the required, accelerator-based infrastructures is still a limiting factor to a wider spread of this type of treatment. In consideration of this, there is an ongoing effort aimed to explore the potential of laser-driven acceleration as a possible cost-saving alternative. Beside a large amount of research aimed to progress laser-based acceleration towards therapeutically relevant parameters, preliminary work has been carried out in the last few years by a number of research groups on the methodology and feasibility of using laser-driven sources for cell irradiation experiments. This work has aimed to establish a procedure for cell handling, irradiation and dosimetry which is compatible with a laser plasma interaction environment, but also to highlight any novel effects related to the ultrahigh dose rates ($>10^9$ Gy/s) that can be delivered to cells by the laser driven ion bursts. Our group has engaged in a number of such experiments, the latest of which have been carried out at the Laboratoire d'Utilisation des Lasers Intenses (LULI) in France.

In this experiment the AG01522 and HUVEC cells were used. The cells were cultured in vitro on a 3 μ m mylar film attached to a metal dish. Both cell lines were exposed to a laser-driven proton beam generated by the interaction of the 150TW LULI2000 laser beam with a gold foil, and suitably dispersed in energy by a dipole magnet. The laser driven proton beam produced a similar dose on cell as used in conventional irradiations (of the order of a Gy), but with a dose rate of many orders of magnitude higher. A number of biological assays (clonogenic assay, immunofluorescence and senescence) were carried out to assess the radiation damage caused to the cells. The talk will discuss the methodology employed and the preliminary results obtained from the analysis of the data.

BLOOD PERFUSION CHANGES IN EXERCISING AND NON-EXERCISING MUSCLE DURING UNILATERAL STATIC EXERCISE USING DIFFUSE CORRELATION SPECTROSCOPY TECHNIQUE

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³*HemoPhotonics S.L., Av. Carl Friedrich Gauss Num. 3, 08860 Castelldefels (Barcelona), Spain*

The mechanisms governing non-exercised muscle vascular response during local exercise, involving a small muscle mass are poorly understood to date. Contractions involving small muscle mass can induce different responses of peripheral resistance in vascular bed of non-exercised muscle.

Therefore the aim of this study was to reveal blood perfusion changes in exercising and non-exercising muscle during unilateral static contraction using NIR-DCS. Five male volunteers were recruited and gave their informed consent. The protocol comprised two parts – for forearm and thigh muscle microcirculation recording, exploiting two light source-detector distances: 1.5 cm for superficial and 2.5 cm for signal acquisition from deeper tissue layer. In order to calibrate signal baseline, arterial occlusion test has been accomplished prior to exercise. Static handgrip test was performed to assess blood perfusion of *m. extensor digitorum* in active and contralateral forearm. NIR-DCS signal was recorded during rest (4 min.), unilateral handgrip contraction (50% MVC, 80 s) and following post-exercise recovery period (5 min.). Knee extension exercise was performed to evaluate blood perfusion of *m.vastus lateralis*. NIR-DCS signal was recorded during rest (4 min), unilateral static knee-extension (50% MVC, 80 s) and following post-exercise recovery period (5 min).

Static exercise induced larger increase of perfusion in deep muscle tissue (distance 2.5 cm) in comparison to superficial. In contralateral extremity a small increase of perfusion during contraction has been observed, possibly due to increased systemic cardiovascular parameters. In active limb, in contrary to contralateral, post-exercise hyperaemia following active hyperaemia has been observed.

We conclude that, NIR-DCS technique utilizing HemoFloMo device, can be used for non-invasive evaluation of skeletal muscle microcirculation during static exercise from different muscle parts at different depths.

Acknowledgements: This study was supported by Project Activity under the Project “LASERLAB-EUROPE III—The Integrated Initiative of European Laser Research Infrastructures III” and “Hemophotonics S.L.”.

TOWARD IN VIVO AND IN SITU TIME DOMAIN REFLECTANCE DIFFUSE OPTICAL TOMOGRAPHY THANKS TO TIME GATED SPAD SYSTEMS

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 Michel Berger¹, Alberto Dalla Mora², Antonio Pifferi² and Jean-Marc Dinten¹

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Near-infrared diffuse optical tomography (DOT) is a medical imaging technique which gives the distribution of the optical properties of scattering biological tissues. To obtain endogenous chromophore features in the depth of a highly scattering medium, a multiwavelength/time domain (MW/TD) optical setup combined to a complex three-dimensional reconstruction algorithm was developed. Experimental measurements were conducted by illuminating a perturbed medium (with an inclusion) with a picosecond near-infrared laser and by collecting, for several wavelengths and multi-positions, the backscattered light via two fibers connected to fast-gated single-photon avalanche diodes (SPAD) and coupled to a time-correlated single-photon counting (TCSPC) system [1,2,3]. The data processing of these TD measurements and those of a known reference (homogenous) medium was performed by using a method based on the Mellin-Laplace transform [4].

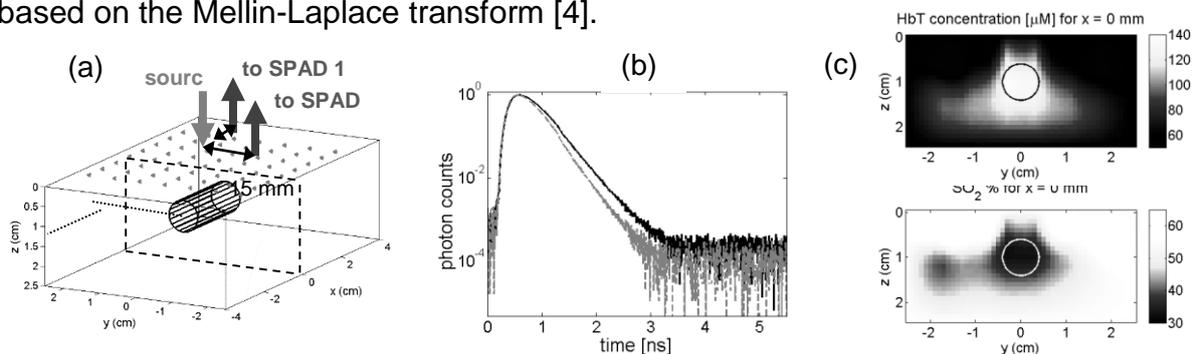


Fig 1: Steps of MW/TD reflectance DOT. (a) Drawing of the phantom and scan set-up. (b) Normalized TD measurements of the reference (black continuous line) and the perturbed medium (grey dotted line). (c) Transversal slices of the 3D reconstructed maps of total hemoglobin (HbT) concentration and oxygen saturation (SO₂) percentage (scale: 50 % threshold of extremum) (Adapted from [3]).

These preliminary phantom and additional numerical experiments show encouraging results to determine chromophore concentrations in depth with a good localization and a good separation of the chromophores. MW/TD reflectance DOT is a promising method to go toward in vivo measurements. The potential clinical applications are breast cancers detection, brain hemorrhages imaging and flaps surgery monitoring.

- [1] Puszka, A.; Di Sieno, L.; Dalla Mora, A.; Pifferi, A.; Contini, D.; Boso, G.; Tosi, A.; Hervé, L.; Planat-Chrétien, A.; Koenig, A.; and Dinten, J.-M. *Biomedical Optics Express* **2013**, *4*, 1351-1365.
- [2] Tosi, A.; Dalla Mora, A.; Zappa, F.; Gulinatti, A.; Contini, D.; Pifferi, A.; Spinelle, L.; Torricelli, A. and Cubeddu, R. *Optics Express* **2011**, *19*, 10735-10746.
- [3] Zouaoui, J.; Hervé, L.; Di Sieno, L.; Planat-Chrétien, A.; Berger, M.; Dalla Mora, A.; Pifferi, A.; Derouard, J. and Dinten, J.-M. *Proc. SPIE 9538, Diffuse Optical Imaging V, 95380C* **2015**.
- [4] Hervé, L.; Puszka, A.; Planat-Chrétien, A. and Dinten, J.-M. *Applied Optics* **2012**, *51*, 5978-5988.

HIGH SPECTRAL RESOLUTION SPECTROSCOPY OF INDIVIDUAL RARE-EARTH-ION DOPED NANOPARTICLES

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Crystals containing rare-earth ions possess many properties that make them attractive for quantum information processing technology. In particular, the narrow linewidths demonstrated on optical [1], electron-spin [2], and nuclear-spin [3] transitions open up exciting possibilities for integrated and hybrid quantum devices. We are seeking to enhance the opportunities for rare-earth-ion quantum technology by developing nanoparticles that preserve the appealing physical properties of bulk crystal systems.

This work concentrates on achieving a sufficiently narrow optical transition linewidth Γ_h to allow single nanocrystals to be strongly coupled to an optical cavity. Recent experiments performed at Chimie ParisTech demonstrated that a Γ_h narrower than 100 kHz could be achieved for Eu^{3+} ions in 60 nm diameter Y_2O_3 nanocrystals at a temperature of 2 K [4]. These measurements were coherent measurements made on powder samples, which contained a large number of nanoparticles, to increase the signal level. The disadvantage of this technique is that any variations in Γ_h due to size, morphology, and surface characteristics of individual crystals are obscured. Accessing the properties of single nanoparticles requires a fluorescence microscope combining kHz frequency resolution and submicron spatial resolution. Such an apparatus has been developed at Lund University to study rare-earth-ion crystals at the single ion level.

The presentation will focus on the spectroscopic techniques developed to probe single nanocrystals and our initial measurements. In particular, solutions for measuring Γ_h on a timescale faster than the fluorescence lifetime of Eu^{3+} (~ 1 ms) will be discussed.

Acknowledgements: The research leading to these results received funding from LASERLAB-EUROPE (grant agreement No. 284464, EC's Seventh Framework Programme), from CIPRIS [People Programme (Marie Curie Actions) of the EU Seventh Framework Programme FP7/ 2007-2013/ under REA Grant No. 287252], and from ANR project RAMACO (12-BS08-0015-01).

[1] Sun, Y. et al., *Journal of Luminescence*, **2002**, 98 (1-4), 281–287.

[2] Wolfowicz, G. et al., *Physical Review Letters*, **2015**, 114(17), 170503.

[3] Zhong, M. et al., *Nature*, **2015**, 517(7533), 1–18.

[4] Perrot, A. et al., *Physical Review Letters*, **2013**, 111(20), 203601.

ULTRAFAST PHOTOEMISSION SPECTROSCOPY STUDY OF THE ELECTRON-LATTICE COUPLING IN THE 2D METALLIC SYSTEM α -Sn/Ge(111)

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⁴Centre Lasers Intenses et Applications, Bordeaux, France

⁵Aix-Marseille Université, CNRS, PIIM UMR 7345, Marseille, France

The α -Sn/Ge(111) surface is a prototypical system where the ground state depends on the interplay between lattice and electron dynamics. This surface is characterized by Sn adatoms regularly located on one out of three T4 sites of the bulk terminated Ge(111) surface.

We present a pump-probe VUV-IR photoemission study of the temporal dynamics (0-20 ps) of the Sn 4d core level spectra under optical excitation (IR 800 nm or 2 μ m). The aim is to unveil the complex electron-lattice coupling mechanism involved in the α -Sn/Ge(111) ($\sqrt{3}\times\sqrt{3}$)R30 $^\circ$ \leftrightarrow 3 \times 3 surface phase transition.

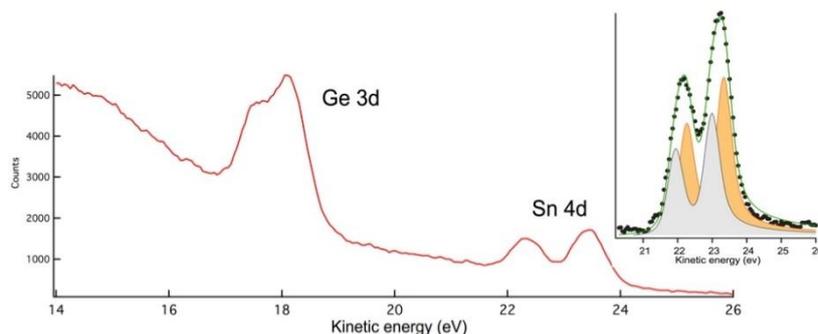


Figure 1: Core level photoemission spectrum collected at the α -Sn/Ge(111).

The photoemission measurements were done at the CELIA facility using the 27th and 29th harmonic of the laser beam produced by the AURORA apparatus.

In Fig. 1 a typical photoemission spectrum collected on the α -Sn/Ge(111) is reported. The Sn 4d core level is characterized by two distinct components as previously reported [2]. These components are assigned to two non-equivalent Sn adatoms where empty and filled surface electronic states are mainly located.

The intensity of the Sn 4d components shows different time evolution when the sample is excited by 2 μ m or 800 nm radiation, pointing to different excitation mechanism for the two wavelengths.

[1] J. M. Carpinelli, et al. *Phys. Rev. Lett.* **1997**, 79, 2859.

[2] J. Avila, et al. *Phys. Rev. Lett.* **1999**, 82, 442; R. I. G. Uhrberg, et al. *Phys. Rev. Lett.* **2000**, 85, 1036; Le Lay et al. *Appl. Surf. Sci.* **1998**, 123, 440.

SIMULTANEOUS TIME-AND-SPACE RESOLVED REFLECTIVITY AND INTERFEROMETRIC MEASUREMENTS OF DIELECTRICS EXCITED WITH FEMTOSECOND LASER PULSES

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Since the first experiments regarding the interaction of fs laser pulses with dielectrics, the fundamental mechanisms of carrier excitation and relaxation have been a subject of intensive research. In particular the relative roles of multiphoton and avalanche ionization in the excitation process under different excitation conditions are still controversial, and experiments sensitive to the real part of the refractive index of the excited material (interferometric) provide carrier population build-up and relaxation times much faster than those sensitive to the imaginary part (reflectivity or transmittance). The lack of spatial resolution in most experimental lay-outs impedes assessing the actual local excitation fluence associated with a given instantaneous carrier density. The role of different carrier excitation/relaxation mechanisms under different excitation levels thus remains unclear. This work aims at clarifying these aspects by analyzing results from simultaneous, spatially-and-temporally resolved measurements of the reflectivity, transmission and refractive index of the excited material (fused silica and sapphire) using different probe wavelengths. The measurements will also be compared to numerical modeling to gain insight into the physical mechanisms involved in the laser-material interaction.

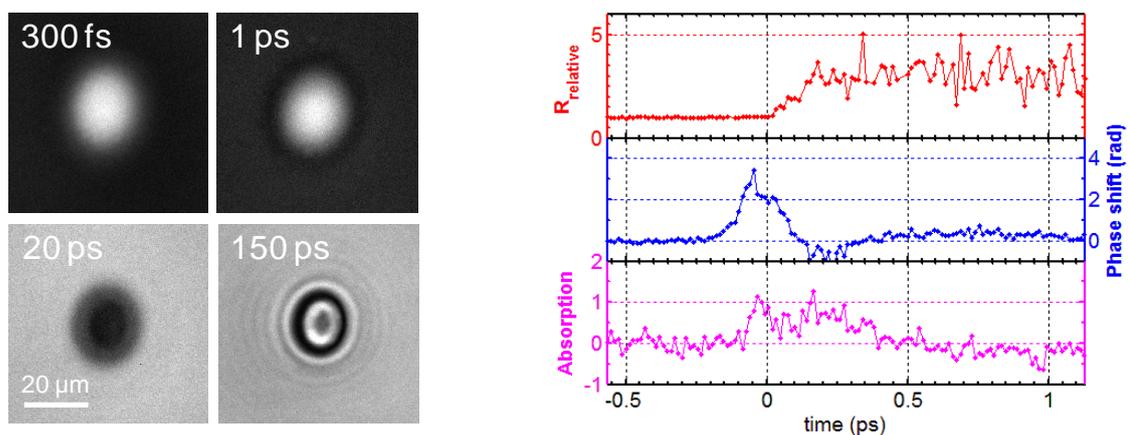


Figure 1: (Left) Images of the reflectivity evolution at 800 nm of sapphire excited with a 400 nm, ~80 fs laser pulse above the ablation threshold. (Right) Illustrative example of the simultaneously measured evolution of reflectivity, phase shift, and absorption of fused silica at 400 nm upon excitation with a 800 nm, ~80 fs, laser pulse above the ablation threshold.

Acknowledgements: This work has been partly funded by LASERLAB-EUROPE (grant agreement no.284464, EC's Seventh Framework Programme) under reference SLIC002014, and the Spanish Ministry Economy and Competitiveness (MINECO, TEC2014-52642-C2-1-R).

SOLID STATE LUMINESCENCE ENHANCEMENT IN DICYANO-DISTYRYLBENZENES: INTRA- AND INTERMOLECULAR CONTRIBUTIONS

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Within the last few decades, aggregation-induced enhanced emission (AIEE) of conjugated organic compounds has drawn much attention, in particular due to applications in optoelectronics [1]. However, the mechanism is not yet fully elucidated, being a complex synergetic process determined by both intra- and intermolecular factors [2,3]. Hence, gaining deeper insights into the AIEE mechanism is highly beneficial towards targeted design strategies for optoelectronic applications. We investigate here structure–property relationships of functionalized dicyano-distyrylbenzenes (DCS, Fig. 1), being a AIEE prototype material. Intra- and intermolecular factors are elucidated independently both in solution and in the solid state through an integrative approach combining steady-state and time-resolved fluorescence and pump-probe techniques with computational methods and photoacoustic spectroscopy (to quantify the deactivation pathways and get detailed knowledge on thermal relaxation) [4]. Our library of molecules with systematic variation of the cyano-substituent position (α/β), the presence of additional alkoxy substituents in the central and/or terminal phenyl rings, allows for systematic tuning of solution and solid state luminescence properties, providing an in-depth understanding of the AIEE mechanism and suggesting design strategies for highly effective functionalized DSB-based materials [4].

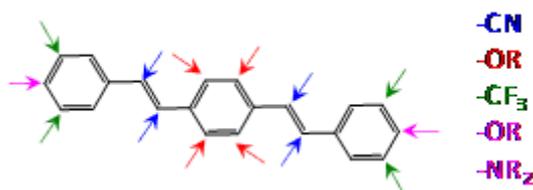


Fig. 1: Functionalization of DSB

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PHOTONIC CRYSTAL SPATIAL (ANGULAR) FILTERS

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The Laserlab project vulrc1970 (the applicant: Politecnical University Of Catalunya (UPC) in Barcelona, Spain, and the host: Laser Research Center LRC in Vilnius University, Lithuania) was devoted to developing of Photonic Crystal (PhC) spatial filters. The basic idea is that the PhCs in addition to their prominent chromatic (frequency) bandgaps, can also display the angular bandgap. We proposed some time ago that these angular bandgaps (or also quasi-bandgaps) can provide the angular i.e. the spatial, filtering [1] (see also recent review on spatial filtering [2], see also Fig.1. for illustration). Such specially designed PhCs can work as stand-alone spatial filters, but, due to extremely small spatial dimensions (thickness is only around 100 μm) can be integrated inside cavities of microlasers.

The collaboration supported by the Laserlab project was mainly devoted to develop the fabrication of such filters, by writing the corresponding structures in glass-like transparent materials by femtosecond pulse technique. I will present the physical principles of such filtering, and will provide the first realizations.

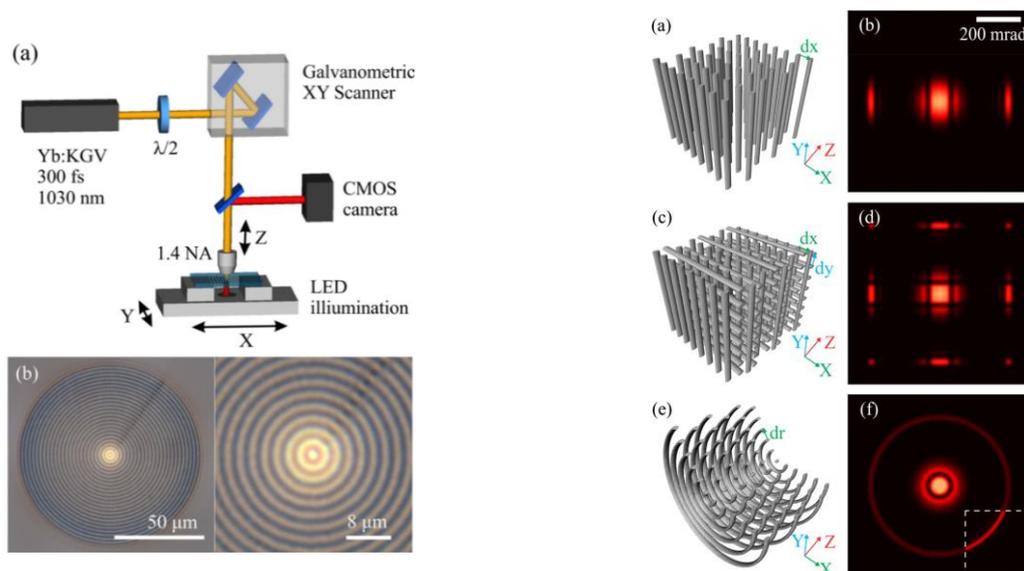


Figure 1: left: laser fabrication of photonic crystal spatial filters; right: different geometries of spatial filters, together with the corresponding light transmission distributions (the far field).

Acknowledgements: We acknowledge Laserlab project vulrc1970 at VULRC, and Spanish project FIS2011-29734-C02-01 of Ministerio de Educación y Ciencia and European FEDER.

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NEAR-INFRARED LASER-INDUCED GENERATION OF HIGHER ENERGY CONFORMERS IN MATRIX-ISOLATED COMPOUNDS

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This contribution will describe experimental facilities existing in the Laboratory for Molecular Cryospectroscopy and Biospectroscopy (at the Department of Chemistry, University of Coimbra), available for scientific and technical training of LaserLab visiting users. In particular, the technique of low temperature matrix isolation and its possibilities in the studies of structure and reactivity of molecules trapped in solid inert gases at cryogenic temperatures (a few Kelvin) will be presented.

Over the past five years, in the frames of the LaserLab training programme, we obtained experimental results providing new insights into the conformational behavior of non-rigid molecules. For several matrix-isolated compounds [1-11], previously unknown higher-energy forms not accessible thermally were generated and characterized experimentally for the first time, using spectroscopic techniques.

Acknowledgements: This work has been supported by the Portuguese “*Fundação para a Ciência e a Tecnologia*”, Research Project PTDC/QUI-QUI/118078/2010 (FCOMP-01-0124-FEDER-021082), co-funded by QREN-COMPETE-UE, and also by The European Community’s Seventh Framework Programme under Grant Agreements No. 228334 and No. 284464. The reported results were obtained in collaboration with foreign colleagues who visited Coimbra as LaserLab trainees: Maciej Nowak, Leszek Lapinski, Anna Halasa, Hanna Rostkowska (Warsaw, Poland), Maria Wierzejewska, Adriana Olbert-Majkut, Justyna Krupa (Wroclaw, Poland), Sonia Melandri (Bologna, Italy). Finally, this research was carried out with participation of the local researchers: Rui Fausto, Cláudio M. Nunes, B. Michela Giuliano (Coimbra, Portugal), whose contribution is also acknowledged.

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PHOTOIONIZED PLASMAS INDUCED BY INTENSE SOFT X-RAY AND EXTREME ULTRAVIOLET PULSES

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In this work investigations of photoionized plasmas were performed using laser-produced plasma (LPP) extreme ultraviolet (EUV) or soft X-ray (SXR) sources with different parameters. The sources were based on three different laser systems with pulse energies ranging from 0.8 J to 500 J and pulse duration 0.2 - 10 ns. Laser plasmas were produced by irradiation of double stream gas puff targets with Xe or KrXe mixture as the working gas. In case of using the high power laser pulses (PALS, Prague) intense SXR pulses were produced. In case of using lasers of low energy, up to 10 J, emission in EUV range dominated the plasma radiation. The ionizing radiation was focused using grazing incidence collectors of different types. The collectors were based on multifoil, ellipsoidal or paraboloidal mirrors optimized for specific wavelength ranges. Photoionized plasmas were produced by irradiation of the gas stream injected in the area of the focused radiation or into the vicinity of the laser plasma.

Different gases were injected into the interaction region, using an auxiliary gas puff valve. Irradiation of the gases resulted in ionization and excitation of atoms and molecules forming photoionized plasmas. Spectra in SXR/EUV range were measured using a grazing incidence, flat-field spectrograph (McPherson Model 251), equipped with a 450 lines/mm toroidal grating and a home made spectrograph based on free standing transmission grating 5000 l/mm. Electron density distributions of photoionized plasmas were measured by laser interferometry, using a femtosecond, Ti:Sapphire laser system. In case of EUV induced PP the electron density was below the detection limit. In case of SXR induced PP electron density distributions were measured for plasmas induced in different gases. In all cases the most intense emission lines were assigned to singly charged ions. Other spectral lines corresponding to doubly, triply and even quadruply charged ions were also recorded. Strong differences between spectra obtained from SXR and EUV induced plasmas are indicated.

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TOWARDS POLARISATION GATING OF HARMONIC EMISSION FROM RELATIVISTICALLY OSCILLATING PLASMAS (HIJ)

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High harmonic generation from intense laser solid density interactions has, over the last decade, demonstrated its clear potential as a next generation source of ultrafast, coherent X-rays. Observations of attosecond phase locking and diffraction limited performance have shown that the exceptional coherence properties of the intense driving laser ($>10^{20}$ Wcm⁻²) can be transferred directly to the extreme ultraviolet (XUV) and X-ray regions of the spectrum with unprecedented efficiency. To date this has been achieved via the formation of a relativistically oscillating mirror (ROM) at the critical density surface (up to 1000's of harmonic orders). Here we present progress towards gating the emission to provide a source of high power attosecond pulses. Furthermore, a novel method of implementing this technique for high power ($\approx >1$ J) laser pulses is presented along with proof of principle experimental results for ROM harmonics¹.

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39 mJ ENERGY, PICOSECOND, 2 μm Ho:YLF CRYOGENIC CHIRPED PULSE AMPLIFIER FOR MID-INFRARED OPCPA

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High energy picosecond pulses at 2 μm wavelength are of importance for developing the next generation of ultrafast laser physics in the mid infrared [1] (mid-IR) spectral region as they permit pumping highly effective mid-IR nonlinear materials efficiently. Amongst many other applications, intense mid-IR few-cycle pulses are key for the development of atto- and zeptosecond science beyond the vacuum ultraviolet and into the soft X-ray regime.

Here, we present a first source operating at 2.05 μm wavelength delivering up to 39 mJ output energy with few picosecond duration and meeting the demanding requirements in stability and beam quality imposed on OPCPA pumps. The system relies on a CPA architecture consisting of a seed laser, a stretcher/compressor pair, a regenerative amplifier and a cryogenically cooled single pass amplifier. The seed laser is based on an amplified, frequency shifted Er: fiber oscillator delivering 4 nJ energy pulses with picosecond duration and 1.2 nm bandwidth centered around 2052 nm. Upon emerging from the fiber laser, the pulses are temporally stretched in a chirped volume Bragg grating (CVBG) based assembly and pulse picked to 100 Hz repetition rate. The first amplifier stage consists of a ring regenerative amplifier relying on a water-cooled Ho:YLF crystal pumped by a 1940 nm wavelength, CW fiber laser. The 5.5 mJ energy pulses delivered by the regenerative amplifier are further amplified in a second amplification stage which is a single pass amplifier based on a cryogenically-cooled Ho:YLF crystal, pumped by a 120 W CW fiber laser at 1940 nm wavelength. Upon amplification, the pulses are temporally compressed in a large aperture CVBG compressor. The laser system delivers optical pulses with up to 39 mJ output energy with 10 ps duration at 100 Hz repetition rate with excellent spatial intensity profile. The system operated over hours daily and entirely alignment free.

These systems find applications as mid- IR OPCPA pumps and are suited to drive inverse Compton scattering experiments.

Acknowledgements: This research was partially supported by LASERLAB-EUROPE III, activity H0021-03

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EFFICIENT LASER PRODUCTION OF ENERGETIC NEUTRAL BEAMS

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Laser-driven ion acceleration by intense, ultra-short, laser pulse has received increasing attention during the past few years, and the availability of much compact and versatile ions sources motivates the study of laser-driven sources of energetic neutral atoms [1-3]. We demonstrate the production of a neutral and directional beam of hydrogen and carbon atoms up to 200 keV per nucleon, with a peak current of 2.7×10^{13} atom/s. Laser accelerated ions are neutralized in a pulsed, supersonic argon jet with tunable density between $1.5 \times 10^{17} \text{ cm}^{-3}$ and $6 \times 10^{18} \text{ cm}^{-3}$. The neutralization efficiency has been measured by a time-of-flight detector for different argon densities. An optimum is found, for which complete neutralization occurs. The neutralization rate can be explained at high areal density ($> 1 \times 10^{17} \text{ cm}^{-2}$) by single electron charge transfer processes.

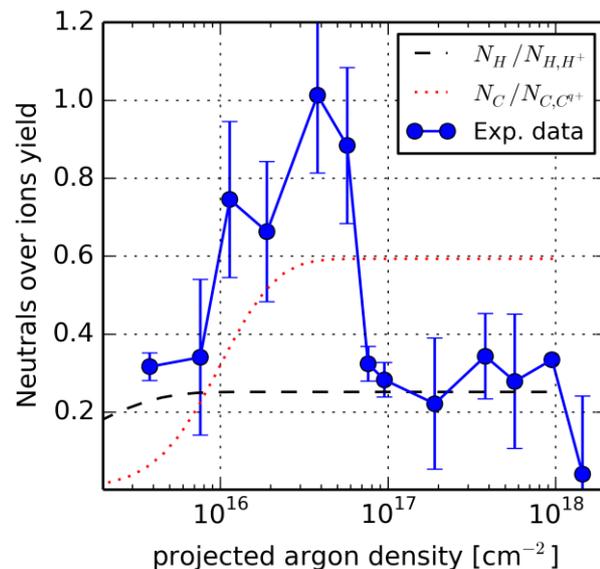


Figure 1: Experimental and computed neutralization efficiency of H and C ions in the range 60 keV/u to 100 keV/u crossing an argon jet.

Acknowledgements: OSEO Project n° I0901001W, ANR ILA, Deutsche Forschungsgemeinschaft (program CRC/Transregio 18) and LASERLAB- EUROPE (EU-FP7 284464, project MBI001857)

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TEMPORAL FLAT TOP SYNTHESIS FOR HIGH ORDER HARMONIC GENERATION

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High harmonic generation (HHG) process comprise the advantages of being a coherent process which enable the production of trains of attosecond pulse in the XUV range. The process itself consists in a 3 steps sequence of coherent events within one half of the optical cycle of an intense and short femtosecond laser pulse [1]. These 3 steps if controlled can be use to probe dynamical process within few hundreds of attosecond. [2]. However experimentally the HHG process is obtained by focusing the laser pulse into a volume of atoms. In order to detect the emission one has to realis phase-matching conditions for constructively build up the emission on a macroscopic level. Therefore the coherent HHG emission detectable is intrinsigly coupled in space and time. So controlling the laser properties in space and time should allowed one to avoid time and space averaging and produce a mascroscopic harmonic emission which mimic the single atom response. Hence enable to scrutinise in-situ the 3 steps sequence of HHG process and use it to probe dynamical process in the target. The synthesis of a spatial flat top laser field has been demonstrated to have an serious impact on controlling the HHG process for producing attosecond train with the same repetition rate as the incoming laser (isolated attosecond scheme) [3].

Hence in this work we have investigated the impact of the temporal synthesis of the laser field on high harmonic generation produced in argon and neon. We found experimental conditions to modify a gaussian temporal envelop towards temporal flat top. We studied its controlled signature in the HHG spectra recorded.

Acknowledgements: All Authors acknowledge Laserlab III CNRS-CELIA002069 for supporting this research. A. Zair and her team acknowledge EPSRC Grant No. EP/J002348/1 for UK support.

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TWO-DIMENSIONAL FREQUENCY RESOLVED OPTO-MOLECULAR GATING OF HIGH ORDER HARMONIC GENERATION

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Probing electronic dynamics in polyatomic molecules poses a significant challenge in high harmonic spectroscopy. Reconstructing the dynamics under study is limited by the multiple degrees of freedom involved in strong field interactions. In this work, we introduce a new measurement scheme that resolves the temporal amplitude and phase of the harmonic emission from the excited molecules.

We record the harmonic spectrum from vibrationally excited N₂O₄ dimers as a function of time. We observe modulations in the harmonic intensity, whose origin has been debated [1]. In addition, high harmonics were generated with a two colour laser field, which enables measurements of the two components of the harmonic polarization. Using a new scheme that we call Frequency-Resolved Opto-Molecular Gating (FROMAGE), we extract the amplitude and phase of the modulation, and thus, of the high harmonic emission without the need for much more elaborate experimental devices.

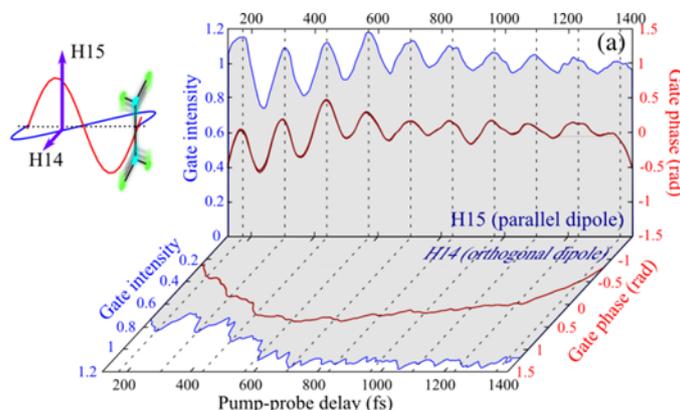


Figure 1: Reconstructed intensity and phase of the modulation for harmonics 14 and 15. The orientations of the molecule and two colour laser fields are shown at left.

Mapping the vectorial properties of the harmonic emission for different molecular orientations will provide insight into ultrafast electronic and vibrational dynamics, particularly for complex molecules where processes initiated by strong laser fields can occur.

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A PLASMA BASED LASER AMPLIFIER

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A plasma-based laser amplifier is a single-pass amplification process that features the interaction of two laser beams, a *pump* (\mathbf{k}_p, ω_p), delivering its energy, and a *seed* (\mathbf{k}_s, ω_s), to be amplified, within a preformed plasma. The mechanism relies on the response of the plasma to an intense laser excitation, in the form of an electrostatic (electronic or acoustic) plasma wave ($\mathbf{k}_{pl}, \omega_{pl}$). This wave fulfills a 3-wave resonant coupling in terms of energy and momentum: $\omega_p = \omega_s + \omega_{pl}$ and $\mathbf{k}_p = \mathbf{k}_s + \mathbf{k}_{pl}$.

Plasma-based laser amplification is considered as a possible way to overcome the technological limits of present day laser systems in terms of damage threshold, by using a plasma as amplifying medium and eventually achieve extremely intense laser pulses.

In a series of experiments we demonstrate the amplification of a picosecond *seed* laser pulse by stimulated Brillouin scattering (SBS, ion acoustic waves) of a *pump* pulse in a plasma, using a fully head-on collision geometry between *pump* and *seed*. A systematic energy transfer of tens of mJ to the seed pulse is recorded. We observe signatures of the transition from linear to self-similar regime of strongly coupled SBS: the strongest amplification is associated with the broadest bandwidth, the shortest pulse duration, and the largest frequency shift. SBS *seed* amplification is also correlated to a strong decrease of the *pump* stimulated Raman backscattering.

XUV EXCITATION FOLLOWED BY ULTRAFAST NON-ADIABATIC RELAXATION IN PAH MOLECULES AS A FEMTOASTROCHEMISTRY EXPERIMENT

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Highly excited molecular species are at play in the chemistry of interstellar media and are involved in the creation of radiation damage in a biological tissue. Recently developed ultrashort extreme ultraviolet (XUV) light sources offer the high excitation energies and ultrafast time-resolution required for probing the dynamics of highly excited molecular states on femtosecond and even attosecond timescales. Here we show that polycyclic aromatic hydrocarbons (PAHs) undergo ultrafast relaxation on a few tens of femtoseconds timescales, involving an interplay between the electronic and vibrational degrees of freedom. Our work reveals a general property of excited radical PAHs that can help to elucidate the assignment of diffuse interstellar absorption bands (DIBs) in astrochemistry, and provides a benchmark for the manner in which coupled electronic and nuclear dynamics determines reaction pathways in large molecules following XUV excitation. It shows possible opportunities for investigating sub-femtosecond dynamics in large molecular species.

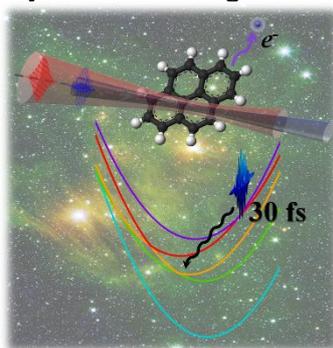


Figure 1: The ultrafast relaxation of excited radical PAHs species is probed in real-time using XUV-IR pump-probe scheme. This reveals general properties of probable diffuse interstellar band carriers.

[1] Marciniak. A et al. Nature Communication, 2015, 6, 7909.

FEMTOSECOND DYNAMICS OF ISOLATED RADICALS IN THE GAS PHASE

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Radicals are important intermediates in combustion processes [1] and in the formation of polyaromatic hydrocarbons (PAHs), a precursor to soot [2]. They are involved in the formation of complex molecules in outer space and in galactic clouds [3], since they are often thermodynamically stable but kinetically instable.

Two radicals were investigated, the benzyl radical and the 2-methylallyl radical. Both radicals were produced from nitrite precursors by pyrolysis and subsequently expanded in a molecular beam. Using femtosecond pump-probe spectroscopy the lifetimes of their excited states were examined via photoelectron spectroscopy and mass spectroscopy.

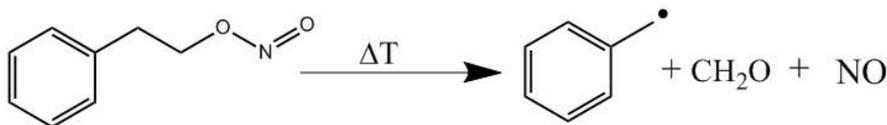


Figure 1: Generation of the benzyl radical via pyrolysis

The benzyl radical was excited in the D₅ state using 266 nm and ionized with either an 800 nm or the BBO-doubled 400 nm probe pulse. The time-resolved mass spectra of the benzyl radical with 800 nm showed two time constants ($\tau_1 = 80$ fs; $\tau_2 = 1.5$ ps), while with 400 nm only one time constant ($\tau_1 = 80$ fs) could be observed.

In the photoelectron spectra with 800 nm a Rydberg fingerprint spectrum was observed, showing the same time constants as before, while the photoelectron spectra with a 400 nm probe pulse showed only one peak with time dependence.

These results confirm the deactivation pathway calculated by the group of R. Mitric (Wuerzburg), in which the initial populated D₅ state decays quickly via the D₄ to the D₃ state (short time constant), which decays more slowly to the D₁ state (long time constant).

In addition preliminary results of the 2-methylallyl radical are presented, which was excited around 240 nm and showed two time constants.

Acknowledgements: Roland Mitric

[1] Miller, J.A.; Kee, R.J.; Westbrook, C.K. *Annu. Rev. Phys. Chem.* **1990**, *41*, 345.

[2] Kohse-Höinghaus K.; Atakan B., Kasper T. *Phys. Chem. Chem. Phys.* **2002** *4* 2056.

[3] Herbst E. *Chem. Soc. Rev.* **2001**, *30*, 168.

ULTRAFAST VIBRATIONS OF SMALL THIOLATE GOLD CLUSTERS STUDIED BY FEMTOSECOND TRANSIENT ABSORPTION SPECTROSCOPY

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The vibrational properties of nanometric size objects have raised large technological and fundamental interest during the last decade. Motivated by the creation of high frequency electrically or optically driven oscillators, the investigation of the acoustic response of small metal nanoobjects is of particular interest. In the past, the validity of the continuous elasticity theory has been shown experimentally for glass-embedded nanospheres as small as 1.3 nm [1].

In this project we investigate the vibrational properties of atomically defined gold clusters (10 to 144 Au atoms), functionalized with different organic molecules in solution using broadband pump-probe spectroscopy with sub-20-fs time resolution. These clusters are in the crossover regime between a solid and a molecule and are ideal candidates to test the breakdown of continuous elasticity theory.

An oscillatory signal has been detected in pump-probe signals for all gold clusters (see Fig. 1). The resulting periods can be attributed to quasi-breathing mode vibrations of the gold atom core. They are found to be in good agreement with complete atomistic simulations [2], and indicate a breakdown of the proportionality between period and diameter predicted by continuous elasticity theory.

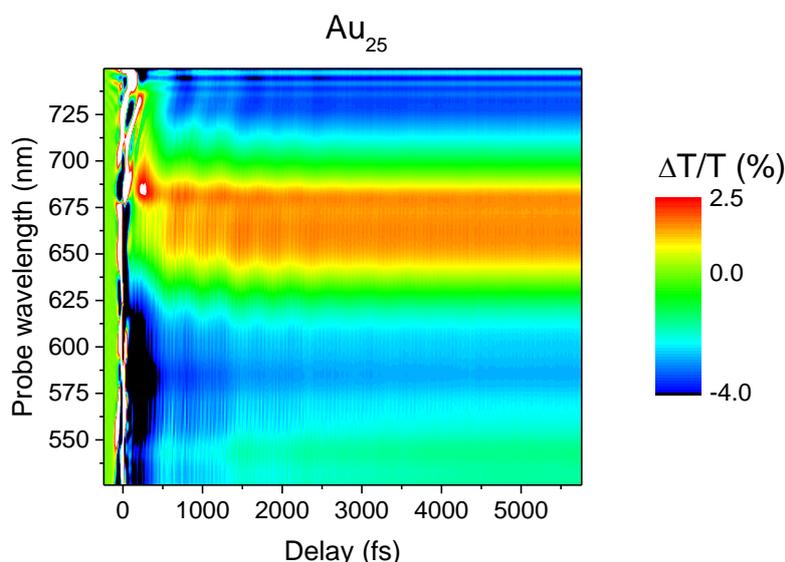


Figure 1: Time resolved pump-probe experiments: transient transmission change $\Delta T/T$, measured in clusters of 25 gold atoms for a pump excitation at $\lambda = 550$ nm.

- [1] V. Juvé, A. Crut, P. Maioli, M. Pellarin, M. Broyer, N. Del Fatti, F. Vallée, *Nano Lett.* **2010**, 10, 1853.
 [2] A. Tlahuice-Flores, R. L. Whetten, M. Jose-Yacamán, *J. Phys. Chem. C*, **2013**, 117, 12191.

LIDT MEASUREMENTS OF OPTICAL COMPONENTS FOR HIGH-ENERGY HiLASE LASERS

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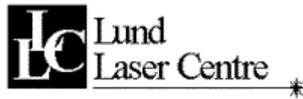
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Laser induced damage threshold is a key parameter for all components in high-power laser system, establishing limits of maximum achievable energy, and consequently average power. To provide reliable and stable laser sources, desirable both in academic and industrial area, involved components has to be tested and meet certain quality criteria. To provide such laser source is the goal of HiLASE project, where the development of scalable kW-class diode pumped solid state pulsed lasers is taking place.

The limiting factor for using and developing laser system and beam distribution for powerful pulsed lasers, delivering peak power at the level of hundred terawatts, is the damage threshold of used components. Although LIDT testing is a part of common procedures conducted by optical component manufacturers, they are not usually suited for extreme conditions regarding HiLASE lasers. Testing of both stock components and prototyped parts developed for HiLASE is then necessary and fortunately, the testing facility is accessible in Vilnius thru the LaserLab activity.

A number of different components were tested for LIDT, namely mirrors, beam splitters and AR windows, in multipulse regime (s-on-1). Components were tested according to their intended use at ps and ns pulse lengths from 10^3 to 10^5 pulses. ISO 21254 standards series compliance of the testing facility further ensures reliability and validity of obtained results.

Tested components were provided both by commercial companies as their stock goods and by various manufacturers as customized optics. Obtained LIDT values were very scattered and we found parts with inferior damage thresholds and with average performance. However, we also tested exceptional components, like dielectric mirror with damage threshold exceeding $100 \text{ J}\cdot\text{cm}^{-2}$ for 10 ns pulse. All results will be used to identify respective components suitable for HiLASE purposes and for further development of beam delivery system optics.



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[CESTA](#)

Centre d'Etudes Scientifiques et Techniques d'Aquitaine, CEA, Le Barp, France



[CLF](#)

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[CLPU](#)

Centro de Laseres Pulsados, Salamanca, Spain



[CUSBO](#)

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University of Szeged, Department of Physics, Szeged, Hungary



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[INFLPR](#)

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LOA

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LULI

Laboratoire pour l'Utilisation des Lasers Intenses, CNRS, Palaiseau, France

MBI

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MPQ

Max Planck Institute of Quantum Optics, Garching, Germany

MUT-IOE

Military University of Technology, Institute of Optoelectronics, Warsaw, Poland

PALS

Prague Asterix Laser System, Institute of Physics, Prague, Czech Republic

SLIC

Saclay Laser-Matter Interaction Center, CEA, Saclay, France

STRATH

University of Strathclyde, Glasgow, United Kingdom

ULF-FORTH

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ULLC

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VULRC

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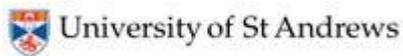


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