



Laserlab Europe

The "**Integrated Initiative**" of European Laser Infrastructures  
in the 7<sup>th</sup> Framework Programme of the European Union



# Laserlab User Meeting

26-27 September  
2013

Marseille, FRANCE



# *Foreword*

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## Foreword by the Meeting Chairs

Laserlab-Europe, in the third phase of successful operation, brings together 30 leading organisations in laser-based inter-disciplinary research from 16 European Member States. Ever since the beginning of Laserlab-Europe, the Transnational Access Programme has allowed more than 1,200 scientists from institutions outside the consortium to have access to the Laserlab facilities to perform their experiments, kindly supported by EC funding. Proposals for Transnational Access are reviewed by an external and independent Access Selection Panel on the basis of scientific merit. To facilitate and foster the exchange of past and potential users with Laserlab laboratories, User Meetings are organized annually at Consortium level. These meetings have become a highly dynamic forum to present access success stories, and also an opportunity for the access providing laboratories to inform about their state-of-the-art facilities.

This time the Laserlab-Europe User Meeting focuses on "**USING LASER SOURCES AND SECONDARY SOURCES AS DIAGNOSTIC TOOLS FOR SCIENCE**" and includes two keynote invited talks given by internationally renowned scientists, 28 oral contributions, a Round Table for discussion of issues of concern to Laserlab-Europe Users and a guided visit of the LP3 laboratories. The meeting is taking place in Marseille, France, kindly hosted by the **Laboratoire Lasers, Plasmas et Procédés Photoniques (LP3)**. Apart from an attractive scientific event, Marseille, as European Capital of Culture in 2013, offers a rich cultural programme to the participants.

We are confident that the assembled programme is of interest to attendees and to the whole Laserlab community and will serve to encourage new collaborations. The Meeting Chairs Marta Castillejo and Marc Sentis are grateful to the Laserlab-Europe facilities for inviting the speakers, to LP3 for kindly hosting the meeting, to the Committee of User Representatives for their organisational support, and particularly to Daniela Stozno and her team for the efficient help during the preparation of the meeting.

The Meeting Chairs

Marta Castillejo, Chair of the Committee of User Representatives of Laserlab-Europe, Instituto de Química Física Rocasolano, CSIC, Madrid, Spain

Marc Sentis, LP3 laboratory, Aix-Marseille University – CNRS, France

# *Schedule*

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# Laserlab User Meeting, Marseille, France, 26-27 September 2013

## Using laser sources and secondary sources as diagnostic tools for science

Status: 4 September 2013

<b>Thursday 26 September</b>		
9h00 - 9h20	Welcome	
<b>Session 1 - Session Chair: Marta Castillejo</b>		
9h20 - 9h50	J. Tisch (invited)	"The generation and application of few-cycle and attosecond light pulses"
9h50 - 10h10	S. Driever	Tunable 1.6-2 $\mu\text{m}$ Near Infrared Few-Cycle Pulse Generation by Filamentation (CELIA)
10h10 - 10h30	S. Ter-Avetisyan	Laser accelerated energetic beams of negative ions and neutral atoms (MBI)
10h30 - 11h00	<b>Coffee Break</b>	
<b>Session 2 - Session Chair: Oldrich Renner</b>		
11h00 - 11h20	L. Lancia	Plasma based laser amplification through Brillouin backscattering (LULI)
11h20 - 11h40	A. Dreischuh	Optical vortices as singular markers in cascaded four-wave frequency mixing process (HIJ-FSU)
11h40 - 12h00	L. Volpe	Absolute reflectivity measurement of spherically bent crystals (CELIA)
12h00 - 12h20	G. Williams	Multiwavelength XUV holography (LOA)
12h20 - 14h	<b>Lunch at MESS</b>	
<b>Session 3 - Session Chair: Rosa Weigand</b>		
14h - 14h30	K. Sokolowski-Tinten (invited)	"Ultrafast processes in laser-excited solids probed by ultrashort X-ray pulses"
14h30 - 14h50	E. Da Como	Broadband ultrafast spectroscopy of organic materials for photovoltaics (CUSBO)
14h50 - 15h10	E. Horvath	Ultrafast spectroscopy on light energy converting nanocomposite materials (USZ)
15h10 - 15h30	P. Balling	Short-pulse laser excitation of dielectric materials: Experiments and modelling (SLIC)
15h30 - 15h50	A. Zair	Dynamical interference signature via High Harmonic Spectroscopy revealing multi-ionisation channel contributions (SLIC)
15h50 - 16h10	F. Lepine	Probing time-dependent decay of multi-electronic super-excited states in PAH molecules (MBI)
16h10 - 16h30	<b>Coffee Break</b>	
16h30 - 20 h	<i>Visit of LP3 laboratory on Campus de Luminy with the possibility for a walk in the Callanques (bus transportation) or visit of MuCEM Museum</i>	
20h	<b>Dinner at Arcenaulx restaurant</b>	

## Laserlab User Meeting, Marseille, France, 26-27 September 2013

### Using laser sources and secondary sources as diagnostic tools for science

<b>Friday 27 September</b>		
<b>Session 4 - Session Chair: Jouko Korppi-Tommola</b>		
9h00 - 9h20	M. Marrocco	Issues on the route to accurate thermometry based on high-resolution vibrational coherent anti-Stokes Raman spectra (LLC)
9h20 - 9h40	M. Lopez-Lopez	Detecting explosives through non-transparent materials using time-resolved Raman spectroscopy; a forensic access project at LaserLaB Amsterdam
9h40 - 10h00	E. Axente	Laser-induced breakdown spectroscopy applied to surface investigation of combinatorial library thin films (LP3)
10h00 - 10h20	M. Modreanu	Investigation of bulk defects in amorphous and polycrystalline HfO <sub>2</sub> thin films (FORTH-IESL)
10h20 - 10h40	Marton/Kisapati	Laser-cleaning of excavated fresco samples and DHSPI testing of model fresco samples for differentiating the photomechanical effects induced by the laser cleaning (FORTH-IESL)
10h40 - 11 h	<b>Coffee Break</b>	
11h00 - 11h20	R. Eason	Shadowgraph imaging in femtosecond laser-induced forward transfer of functional materials (LP3)
11h20 - 12h20	Round table discussion between users, Access Board members and lab directors. Chairs: Rosa Weigand, Jouko Korppi-Tommola, Sylvie Jacquemot, Didier Normand	
12h20 - 14h	<b>Lunch at UNM restaurant</b>	
<b>Session 5 - Session Chair: Istvan Földes</b>		
14h00 - 14h20	R. Laasner	Excitonic absorption saturation with femtosecond UV laser pulses (VULRC)
14h20 - 14h40	G. Tallents	Probing energy transport with EUV lasers (LASERIX)
14h40 - 15h	T. Balciunas	Direct carrier-envelope phase control of an amplified laser system (VULRC)
15h - 15h20	A. Green	Ultra-fast pulsed neutron sources driven by high intensity laser pulses (LULI)
15h20 - 15h40	A. Klisnick/A. Le Marec	Spectral and coherence properties of the PALS X-ray laser (PALS)
15h40 - 16h	<b>Coffee Break</b>	
<b>Session 6 - Session Chair: Marc Sentis</b>		
16h - 16h20	U. Chaulagain	An experiment to study Radiative Shocks relevant to astrophysics at PALS Facility (PALS)
16h20 - 16h40	T. Chodukowski	Investigation of energy transfer from iodine laser shock wave generated in solid target relevant to shock ignition (PALS)
16h40 - 17h	D. Denis-Petit	Nucleus-electronic cloud coupling in plasma: the case of <sup>84</sup> Rb (GSI PHELIX)
17h - 17h20	A. Puszka	Diffuse Optical Tomography with fast-gated SPADs (CUSBO)
17h20 - 17h40	F. Rosmej	Characterization of hot electrons in dense plasmas by high-resolution X-ray imaging near 8 keV (PALS)
17h40 - 18h00	E. Kroupp	High resolution x-ray spectroscopy of laser-produced solid-density plasmas (HIJ-FSU)

***Session 1***

***Session Chair: Marta Castillejo***

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# **The generation and application of few-cycle and attosecond light pulses**

**J.W.G. Tisch, Physics Department, Imperial College London, UK**

After a general introduction to Attosecond Science, this talk will describe recent efforts to develop new sources, including attosecond pulses in the VUV, spectrally isolated attosecond pulses from resonantly enhanced HHG in plasma plumes and enhanced HHG driven by sculpted waveforms formed by multi-colour field synthesis. The energy scaling and CEP stability of few-cycle pulse generation using hollow fibre pulse compression will also be discussed. A recently demonstrated all-optical technique using HHG to characterise arbitrary femtosecond waveforms will also be described.

# Tunable 1.6-2 $\mu\text{m}$ Near Infrared Few-Cycle Pulse Generation by Filamentation

S. Driever<sup>1</sup>, D. Bigourd<sup>1</sup>, N. Federov<sup>2</sup>, M. Cornet<sup>1</sup>, M. Arnold<sup>1</sup>, F. Burgy<sup>2</sup>, S. Montant<sup>2</sup>, S. Petit<sup>2</sup>, D. Descamp<sup>2</sup>, E. Cormier<sup>2</sup>, E. Constant<sup>2</sup>, and A. Zair<sup>1</sup>

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## Abstract

We present results on tunable few-cycle laser pulses generated in the near infrared region obtained by filamentation in a krypton cell combined with group velocity dispersion compensation in fused silica. We obtain a spectral broadening of a factor  $\sim 2-3$  over the entire spectral domain studied. The central wavelength is tuned from 1.6  $\mu\text{m}$  to 2  $\mu\text{m}$  via an optical parametric amplification source. In optimum experimental conditions, where the input central wavelength is set to 1.7  $\mu\text{m}$ , 1.8  $\mu\text{m}$  and 1.9  $\mu\text{m}$ , the achieved spectral broadening covered  $\sim 300$  nm. Using group velocity compensation in bulk fused silica, we obtain near infrared output pulses as short as 2-3 optical cycles with 200  $\mu\text{J}$  energy per pulse. This near infrared filamentation tunable few-cycle pulse source is an important achievement for strong field physics applications such as attoscience, where wavelength scaling has an important effect.

## Background

In recent years several groups started pushing the center wavelength for high harmonic generation further and further to the near IR. A center wavelength between 1  $\mu\text{m}$  and 2  $\mu\text{m}$  bares significant advantages compared to 800 nm. The cut-off energy of the high harmonics scales with  $\propto \lambda^2$ . On the other hand the process gets less efficient the longer the wavelengths ( $\propto \lambda^{5.5}$ ). Hence going to few cycle pulses in this wavelength range is highly favorable.

This was tackled theoretically and experimentally [1]-[8]. Several experimental approaches have been taken in order to achieve this. Sophisticated OPA and OPCPA systems were used by [1] and [2] with subsequent filamentation post compression at 2  $\mu\text{m}$  and 1.5  $\mu\text{m}$ . The hollow core fiber compression was employed at 1.8  $\mu\text{m}$  [4]. To our knowledge there is no study to bridge the gap between these wavelengths using filamentation or their tunability.

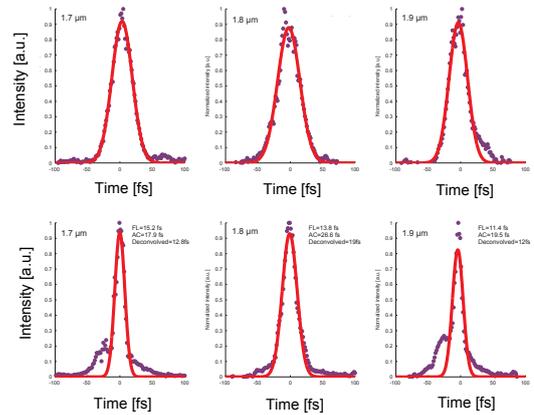
## Experiment

The experimental setup comprised a 800 nm Ti:sapphire laser which was used to pump a commercial OPA system which was tunable from 1.6 to 2  $\mu\text{m}$ . The output was focused loosely into a 4 bar statically filled Krypton cell. We managed to achieve tunable broadening for these wavelengths. In best experimental conditions it was possible to achieve about 300 nm bandwidth for selected wavelengths. A second order autocorrelation of 1.7  $\mu\text{m}$ , 1.8  $\mu\text{m}$  and 1.9  $\mu\text{m}$  showed compression of a factor 2-3 down to the few-cycle regime, see Fig. 1. This compression was achieved by employing the negative GVD of fused silica at these wavelengths. The input energy varied from 460 to 550  $\mu\text{J}$  for the different wavelengths resulting in about 200  $\mu\text{J}$  in the filament.

## Conclusion

Filamentation proved itself to be a versatile tool for post-compression with GVD compensation by fused silica in the near IR. Further studys will be

conducted in April to deepen the understanding and application in HHG.



**Fig. 1.** SHG-Autocorrelation traces for the input idler pulse (upper panel) and the output filamentation compressed pulse (lower panel)

## Acknowledgement

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## References

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## **Laser accelerated energetic beams of negative ions and neutral atoms**

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Beams of energetic negative ions and neutral atoms (oxygen, carbon and hydrogen) are obtained from liquid (water and ethanol) spray targets irradiated by high-intensity ( $5 \times 10^{19}$  W/cm<sup>2</sup>), ultra-short (40 fs) and high contrast ( $< 10^{-8}$ ) laser pulses. Absolute spectra are measured with a Thomson spectrometer enabled for measuring both positive and negative ions. The generation of a beam of energetic neutral atoms was confirmed with CR-39 detector and their spectral characteristics were measured using time of flight technique. Generation is ascribed to electron-capture and -loss processes in the collisions of laser accelerated high-energy protons with a cloud of droplets. The same method can be applied to generate energetic negative ions and neutral atoms of different species.

The quantitative characterisation of the ion yield suggests that the scenario leading to negative ion and neutral atom emission consists of two steps: acceleration of positive ions at the laser focus, followed by interaction of the ions with the remainder of the spray, resulting in the modification of accelerated ion charge state and spectral distribution. Details of the experimental findings and additional evidences of the proposed acceleration scenario will be presented.

The observation opens new possibilities for creation of efficient and compact sources of energetic different negative ion species and neutral atoms.

This research was supported by LaserLab Europe proposal MBI001668.

## ***Session 2***

***Session Chair: Oldrich Renner***

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## Plasma based laser amplification through Brillouin backscattering

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The development of plasma based devices to amplify and focus light at high intensities is one of the most promising steps in the production of ultra-high intensity lasers pulses. Light amplification in a plasma indeed opens the possibility to overcome the limits of present day solid-state based technology, which is subject to damage in manipulating high power densities and heat loads<sup>1</sup>. A plasma amplifier is based on the energy exchange between two laser pulses, a pump and a seed, within a plasma that acts as the coupling and amplifying medium. The kind of plasma response to electromagnetic radiation (i.e. electrostatic modes of the plasma) characterizes the amplification scheme, and the first proof of principle experiment using Brillouin backscattering in the regime of strong coupling (sc-SBS)<sup>2</sup> was performed successfully at LULI. A factor 30 of relative amplification in energy of a few-mJ 400fs seed beam, was obtained<sup>3</sup>. This interaction and coupling scheme has been improved by setting a more advanced (head-on) geometry allowing a major and more efficient exchange of energy between the laser pulses. Plasma conditions have also been improved in order to avoid sources of energy losses. An optimization of the energy exchange based on the shaping of the plasma density profile and the choice of the interaction point of the pulses within the plasma will be performed in an upcoming experimental campaign, as motivated by recent numerical studies. Results obtained from this upgraded scheme will be presented.

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<sup>1</sup> B. Stuart et al., Phys. Rev. Lett. 74, (1995) 2248

<sup>2</sup> A. A. Andreev et al., Phys. Plasmas 13, 053110 (2006).

<sup>3</sup> L. Lancia et al., Phys. Rev. Lett. 104, (2010) 025001

# Optical Vortices as Singular Markers in Cascaded Four-Wave Frequency Mixing Process

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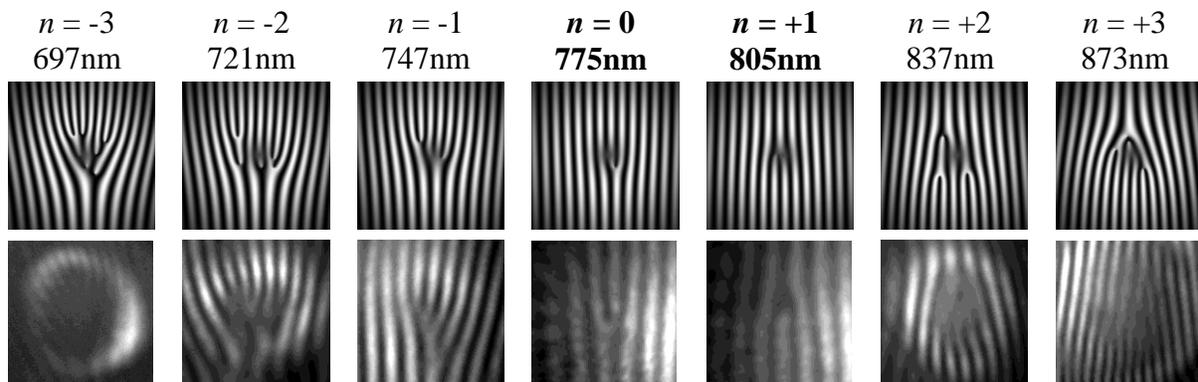
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As a ubiquitous phenomenon in nature, vortices have become an interesting research topic in many fields of physics, which span from the most common observation of fluid-vortices to Bose-Einstein condensates. In the optical domain, vortices are identified as helical phase profiles within a light beam. The central singular point of this helix possesses no defined phase and therefore the intensity must vanish, leading to a characteristic toroidal intensity profile [1]. Such beams carry photon angular momentum, which can also be transferred to matter [2]. This angular momentum is usually referred to as the topological charge (TC)  $m$ , which corresponds to the total phase change  $2\pi m$  over the azimuthal coordinate  $\varphi$ . Of particular interest are nonlinear processes involving such vortex beams, as topological charge constitutes an additional constant of motion (besides photon energy, parity etc.). Up to now experimental evidence of cascading has only been shown in terms of modulated intensity profiles [3], or nonlinear interaction has only been observed up to first cascading order [4].

In this talk we describe broadband cascaded mixing (up to 3<sup>rd</sup> order and in excellent agreement with theory) of vortex beams in a Kerr medium. The topological charge generated from any two spectral components within the bandwidth of the pumping light follows a law analogous to the one for photon energy. The nonlinear generation process, although not phasematched, is efficient enough to allow for observation of vortices in the spectral satellites over a bandwidth  $>200\text{nm}$ . For the first time, the TC of the generated vortex cascade is measured. This constitutes the first measurement of topological charge for a *multiply cascaded* four-wave mixing process with vortex beams.



Theoretical (top row) and experimental (bottom row) interferograms between a collimated Gaussian reference beam and the vortex beams of different order for the four-wave mixing of two pump vortex beams ( $n=0$  and  $n=+1$ ) of opposite TCs  $m = \pm 1$ .

## References

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## Absolute reflectivity measurement of spherically bent crystals

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Spherically bent Bragg crystals are commonly used to image the x-ray fluorescence signal in a very narrow spectral range. They are commonly used in fast electron transport experiments to get a spatial information about a fast electron beam passing through a fluorescence tracer. It is also interesting to quantify the absolute number of photons imaged to deduce the number of fast electrons. For that, a characterization of the crystal efficiency is needed.

Here we present experimental results concerning the “absolute measurement of spherically bent crystals reflectivity”. The experiment, supported by LASERLAB, has been performed in the Eclipse laser facility at CELIA laboratory in Bordeaux. This laser can provide at a high repetition rate (10 Hz) very short laser pulses (40 fs) focused in a 8  $\mu\text{m}$  FWHM focal spot reaching peak laser intensity on target of the order of  $10^{18}$  W/cm<sup>2</sup>.

Different crystals have been used to collect the 8.048 keV  $K_{\alpha}$  radiation coming from the interaction between a laser-generated relativistic electron beam with thin (10  $\mu\text{m}$ ) copper foils. The collected radiation has been reflected into a CCD camera placed at the image plane of the crystal. The same detector has been used to collect photons in the range of 5 to 20 keV coming from the same target in single hit mode. The comparison between the two acquisitions permit us to perform an absolute estimation of the crystal reflectivity. In addition, for both acquisitions we have used an additional CCD camera in single hit mode as reference. Experiment results will be presented in comparison with equivalent results obtained elsewhere and published in literature as well as with the theoretical predictions.

# Multi-wavelength holography using laser generated high-order harmonics: towards sub-micron, atto-second timescale holograms

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Using high order harmonics generated by intense laser interaction with a gas as a source of short x-ray pulses has proven extremely promising in the field of sub micron holography. For the purpose of ease of image reconstruction, generally only a single harmonic wavelength with a narrow bandwidth is selected and used for hologram recording. However, the mixing of multiple harmonics in time has produced the shortest pulses of coherent light in the laboratory to date, allowing for example; the mapping in time of electron motion in atoms and molecules. Using similar multi-colour pulses to record holograms is the next step in atto-second imaging, yet challenges arise in the re-construction of the image when the interference pattern consists of multiply closely spaced wavelengths interfering with each other on the detector plane. We propose two experimental configurations that allow re-construction by the simple Fourier transform approach, overlapping and separating the images by specific placement of the reference.

## ***Session 3***

***Session Chair: Rosa Weigand***

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## Ultrafast processes in laser-excited solids probed by ultrashort X-ray pulses

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Ultrafast pulsed excitation of solids allows to create states of strong electronic excitation and high temperature and pressure. Subsequent to the initial deposition of energy a complex chain of secondary relaxation processes can lead to structural dynamics on very rapid time-scales, and often along unusual, non-equilibrium pathways. The advent of ultrafast diffraction techniques has made it possible to directly follow these structural changes with atomic scale spatial and temporal resolution.

In this contribution I will present some results from our recent work in this field where we used fs X-ray pulses from lab-based (laser-produced plasma) as well as accelerator-based (XFEL) sources to investigate the structural response of solid materials after intense fs laser excitation.

Examples to be discussed include the generation and relaxation of large amplitude coherent optical phonons in Bi where we observe complete softening of the excited A<sub>1g</sub>-mode at sufficiently high excitation fluences. This gives clear indication that the material makes a transition to a transient state of higher symmetry through a reversal of the Peierls-distortion that defines the equilibrium structure of non-excited Bi.

We will also discuss experiments on the switching dynamics in so-called *phase change materials* (PCMs). These materials exhibit rapid and reversible phase transitions between an amorphous and a crystalline state, which can be triggered by short light or electrical pulses. Since the structural changes are associated with large differences in the electronic and optical properties of the two phases, PCMs are widely used for optical data storage as well as in electronic memory applications. We have applied time-resolved X-ray scattering using the XPP-instrument of the Linear Coherent Light Source (LCLS), the worlds first hard X-ray FEL, to directly probe the structural dynamics in various PCMs after laser irradiation over an extended time range from fs to  $\mu$ s. We find that with fs excitation both transitions (amorphous-to-crystalline and crystalline-to-amorphous) involve melting of the material. Depending on the excitation strength melting can occur very fast on a sub-ps time-scale as a non-thermal process driven by the strong laser-induced electronic excitation. However, it takes ns up to tens of  $\mu$ s for the material to reach its final amorphous or crystalline state. These time-scales imply purely thermal mechanisms, determined by the nucleation and growth kinetics for a given material and sample geometry.

# Probing charge carriers in carbon materials with optical pulses

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## Abstract

In optoelectronic devices charge carriers are converted into light by forming excitons, for example in lasers and LEDs, or light is absorbed and excitons are converted into charge carriers when considering photodiodes and solar cells. Optical spectroscopy is a formidable experimental tool to understand light matter interaction and exciton dynamics, but less common for probing charge carriers.

In this seminar, I will present recent experiments based on mid-infrared optical spectroscopy (1.3-0.3 eV photon-energy) aiming to detect and study charge carrier dynamics in graphene materials and conjugated polymers. In the case of graphene, infrared optical pulses are used to understand carrier cooling after photoexcitation<sup>[1]</sup> and measure the hot carrier lifetime<sup>[2]</sup>. For conjugated polymers, the photoinduced generation of polaron-pairs in donor acceptor copolymers is addressed considering the role of molecular structure<sup>[3]</sup> and excess photon energy<sup>[4]</sup>.

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## Ultrafast spectroscopy on light energy converting nanocomposite materials

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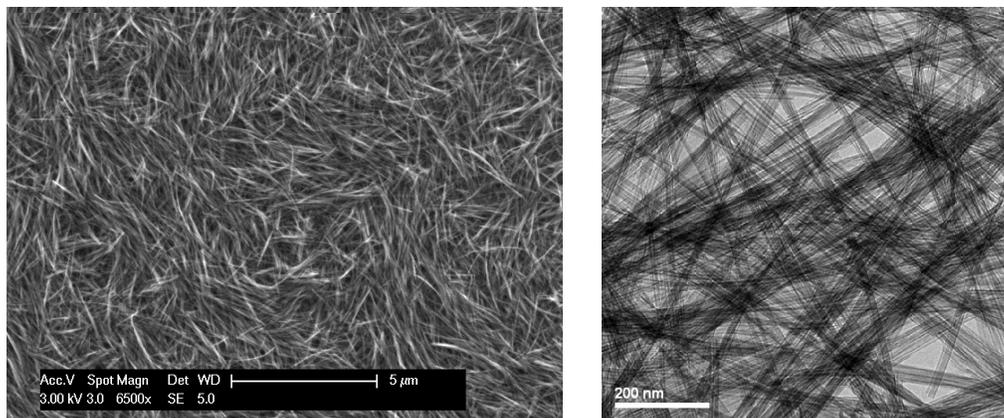
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Application of nanomaterials has been impressively emerging both in research laboratories and at industrial levels. Among these materials those composites, which can be excited by light became in the focus of attention in many laboratories, because of the possibility to convert the light energy very efficiently. These new types of materials open large numbers of new generation applications in energy converting devices, some have been already realized, while others remain for the future.<sup>1-3</sup> There is no debate that the biggest challenge is to use them in energy harvesting systems (like photovoltaics), or optoelectronic devices (as integrated optical elements in optical switches, sensors, imaging equipments, etc.). The most considerable issue of the recent researches is the use of bio-nanocomposite materials, because the biological part is extremely sensitive and specific to given reactions.

Although several light sensitive new types of “nanodevices” have been described recently in the literature, the main problems, like efficiency (quantum and/or energy yield, absorption cross section), stability, reproducibility, recovery of the system, etc., have remained still unanswered.

The work presented here focuses on the characterisation of the kinetic, thermodynamic and structural requirements of light sensitive nanomaterials (inorganic nanowires and bio-nanocomposites) by fast (sub-picosecond/nanosecond) kinetic spectroscopy, which is available in the TeWaTi laboratory of USZ.



SEM and TEM images of photoactive titanium oxide nanowires

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# Short-pulse laser excitation of dielectric materials: Experiments and modelling

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A proper description of ultrashort-pulse laser excitation of dielectric materials is complicated by the strong interplay between the material excitation and the light propagation: during the laser irradiation, a large fraction of the electrons in the valence band are excited to the conduction band inducing large changes to the optical properties of the material. Essentially, the material changes from a transparent dielectric to a highly-reflective and opaque material within the laser pulse duration. This means that a model for short-pulse excitation must account for this interaction, for a recent review, see Ref. 1.

In the current presentation, we will describe a comparison between experimental results and one approach to modelling highly-excited dielectrics. The model is based on a multiple-rate-equation model describing the strong-field excitation of electrons to the conduction band (in a Keldysh description [2]) and the subsequent heating of electrons in the conduction band by inverse Bremsstrahlung followed by collisional excitation of additional electrons from the valence band [3]. In Ref. 4, this model was extended to account for the propagation of the light pulses, which allows the model to be used for predictions of bulk phenomena, such as material-removal (ablation) rates.

Experimental investigations comprise irradiation of well-defined (single-crystal) dielectric materials. The self-reflectivity of the light is measured, and the geometry of the laser-ablated structures is measured by atomic-force microscopy (AFM) [5,6]. In recent investigations, single-shot studies on single-crystal sapphire samples combine time-resolved spectral interferometry with time-resolved reflectivity and ablation-rate measurements over a large span of intensities. The experimental observations are used to test the model and extract relevant material parameters as, e.g., electron scattering rates in the highly-excited material.

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# Dynamical interference signature via High Harmonic Spectroscopy revealing multi-ionisation channel contributions.

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We investigate the attosecond electron dynamic involved in N<sub>2</sub>O and CO<sub>2</sub> after strong field ionisation by characterizing the emitted high order harmonic generation (HHG) in amplitude and phase. We find remarkable differences between the signal detected from these two molecules with comparable electronic structures. One possible reason for these observed differences is the contributions of several of ionisation channels to the HHG. Advanced theoretical modelling of both structural and dynamical effects will be given in order to interpret the experimental observations.

High Harmonic Spectroscopy has attracted tremendous attention recently due to its ability to investigate molecular properties with attosecond and Ångström resolutions [1–3]. The power of High Harmonic Generation (HHG) for probing dynamical process relies on its inherent ‘pump-probe’ scheme. Indeed, HHG can be intuitively understood as the subsequence of 3 steps: tunnel ionization of an electronic wave packet (EWP) originally bound to the molecule (pump), acceleration of the released EWP by the strong laser field (time-frequency mapping allowing accessing attosecond dynamical processes) and recombination (probe) to the core with emission of a burst of XUV coherent light (observable then detected) [4]. Hence the HHG emission encodes fruitful information on the generating system itself, including molecular structure and intra-cation dynamics. A typical feature appearing in the harmonic spectra is a minimum (and a related ‘jump’ in spectral phase) that encodes the structure of the involved bound wave-function, since the emissions from the different orbital lobes interfere destructively, referred to as ‘structural interference’. However, spectral minima can also result from the destructive interference between different ionization channels of comparable contributions to the HHG signal, referred to as ‘dynamical interference’. In order to disentangle which of these two scenarii is likely to occur, one needs to characterize the variations of minima and the correlated phase jump when changing the generation conditions (molecular orientation, laser intensity...). Therefore we perform both spectral amplitude and phase measurements for aligned N<sub>2</sub>O and CO<sub>2</sub> molecules. As expected we observe a spectral minimum and a corresponding phase jump in both molecular system which position in harmonic photon energy is dependent upon the laser intensity, indicating a ‘dynamical interference’.

Comparing the N<sub>2</sub>O case to the one of CO<sub>2</sub> in the same generation conditions, we noticed differences for a given laser intensity: 1/ The phase jump observed is rather steeper in the case of N<sub>2</sub>O than CO<sub>2</sub>; 2/ the phase jump in the case of NO<sub>2</sub> has a greater amplitude than for CO<sub>2</sub> and is of opposite sign.

Although we suspect that in both case the dynamical interference – due to multiple ionization channels – is responsible for this phase jump, we have investigated further theoretically if these differences can be explain as a HHG dependences upon the relative weight of the different orbitals involved in the process. The obtained results allow us to explain how the measurement is related to the multiple orbital contributions and more importantly, to potentially extract the relative ratio between them. Furthermore, controlling the molecular alignment allows us to ‘pre-select’ which orbitals is dominantly involved in the HHG process and additional control of the laser intensity allows us to trigger the relative ratio of pre-selected orbitals’ contribution to the HHG signal. As a good example of such an achievement, our experimental data and theoretical analysis reveal that by changing the laser intensity in the interaction region, we were able to select the dominating orbital to N<sub>2</sub>O harmonic emission from HOMO to HOMO-1.

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# Probing time-dependent decay of multi-electronic super-excited states in PAH molecules

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PAH molecules are very important species in nature, their abundance in space lead to their suspected role in the appearance of complex (bio-)molecules and life. The role they play as a source of electrons is also crucial as electrons are known to induce major damages in biomolecules. As a consequence, the reactions induced by excitation by XUV radiation (present in space for instance through solar winds), is crucial for understanding the evolution of these species in astrophysical chemistry. In this photon energy range, the role played by many-electron excitation remains unclear. One of the important advances made in laser technology relies on the ability to synthesize short femtosecond or attosecond XUV pulses. Nowadays, such pulses can be used in pump-probe experiments to investigate ultrafast dynamics of highly excited states and possibly reaching super-excited states. These states are especially interesting because they deal with exchange-correlation and non Born-Oppenheimer dynamics that remain extremely difficult to comprehend within modern many-body theories. Recently, several important experiments have addressed the role of multi-electronic effects in small molecules such as H<sub>2</sub>, N<sub>2</sub> or O<sub>2</sub> showing that double excited states, autoionization or non-Born-Oppenheimer dynamics occurring in the attosecond or femtosecond time domain, can be probed in real-time using ultrashort XUV pulses. The role that such states play in more complex molecular systems remain unclear and so far experimentally unexplored in real-time. In carbon based molecules, shake-up states have been identified with the help of electron spectroscopy even at surprisingly low photon energy. In PAH, theoretical investigations have supported the existence of such states at photon energy above 10 eV. Recent investigations with synchrotron radiation have demonstrated the existence of new ionization mechanisms for these molecules.

We report the first observation of ultrafast relaxation of highly excited electronic states in PAH molecules following the absorption of XUV photons. Molecules are excited with an ultrashort XUV pulse creating a multi-electronic super-excited state above the ionization threshold. This state is strongly coupled to the ionization continuum. The decay is subsequently probed by a second short IR pulse, revealing a dynamical process faster than 30 fs. This lifetime evolves with the size of the molecule. This mechanism appears as a major relaxation channel of the molecule and was observed for several PAHs, indicating its probable universal role in the relaxation pathways of XUV photo-excited PAHs.

## ***Session 4***

***Session Chair: Jouko Korppi-Tommola***

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# Issues on the route to accurate thermometry based on high-resolution vibrational coherent anti-Stokes Raman spectra

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Coherent anti-Stokes Raman scattering (CARS) is a non-linear optical technique that is at the base of thermometric and concentration measurements in harsh environments [1]. It has become very popular in combustion diagnostics, especially for the potential of taking real-time (i.e, single laser-shot) data on specific molecules that are Raman active (diatomics, mainly). It works thanks to the statistical distribution of the molecular population among the vibrational and rotational energy levels in the ground electronic state and three laser beams are necessary to generate the CARS signal. Two (pump and Stokes lasers) select the Raman modes and the third laser beam probes the Raman response.

In this work, we focus on problems that arise in measurements of vibrational spectra obtained from an experimental set-up mounted at the Lund Laser Centre (LLC) and meant for high-resolution performances with nanosecond lasers. The latter consist of degenerate pump and probe lasers operated in single-mode configuration and coupled to a broadband Stokes laser. The detection is ensured by a relay lens system [2]. It enhances the spectral dispersion to about  $0.02 \text{ cm}^{-1}/\text{pixel}$ .

Technical and physical aspects are involved. Most interestingly, we observe signs of a puzzling spectral oscillation that alters the thermometric measurements. The effect manifests itself in an oscillation of a well-defined frequency that is mixed with the main CARS spectral intensity of the gas under study. To identify the possible causes of this effect, we present a few tests that show the independency from the gas or any other trivial causes associated with the experimental set-up. A method to analyze the oscillation is given and, putting that into practice, a good thermometric evaluation can be achieved. This is applied to the typical nitrogen CARS spectrum measured at room temperature and atmospheric pressure. A possible explanation is given in terms of the temporal treatment of the pulses generated by the nanosecond lasers of our set-up [3, 4].

The analysis developed here has also some further consequences for the role played by other laser characteristics (for instance, pulse asymmetry) that appear to be not negligible for the correct interpretation of the Raman linewidth, whose value is crucial for accurate measurements at high temperatures.

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**“Noninvasive detection of explosives through strongly scattering plastics using time-resolved Raman spectroscopy; a forensic access project at LaserLaB Amsterdam”**

María López-López

The detection of explosives concealed behind strongly scattering materials is becoming a priority in terms of Homeland and International Security, and has attracted particular attention in recent years due to a heightened threat of terrorism. This challenge requires noninvasive analytical techniques for identification without having to manipulate the package. Time Resolved Raman Spectroscopy (TRRS) is a Raman technique that allows the discrimination of surface Raman photons and Raman photons from deeper within a sample on the basis of their distribution in time as they travel through an translucent material and are diffusely scattered and delayed. In this context, experts in the analysis of explosives from the University of Alcalá (Spain) and experts in the use of TRRS from LaserLaB Amsterdam, collaborated to study the possible application of this technique to the analysis of hidden explosives.

A TRRS spectrograph composed of a pulsed laser (3 ps) and a delayed, relatively short detector gate width (250 ps) produced by an intensified charge-coupled device (ICCD) detector were used. The explosive related compounds 2,4-dinitrotoluene, 2,6-dinitrotoluene, akerdite II, diphenylamine, and ethylcentrilte were detected through several mm of translucent polymers or plastic packaging materials (polytetrafluoroethylene (PTFE), polyoxymethylene (POM), polyethylene (PE), polystyrene (PS), and polyvinyl chloride (PVC)). Additionally, a comparison between TRRS and Spatially Offset Raman Spectroscopy (SORS) techniques for the analysis of hidden explosives was studied highlighting the advantages and drawbacks of both techniques. The study provided a new analytical tool for the non-invasive detection of explosives in the security and forensic fields.

# Laser-induced breakdown spectroscopy applied to surface investigation of combinatorial library thin films

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## ABSTRACT

Laser-induced breakdown spectroscopy (LIBS) has demonstrated to be a versatile tool for quantitative analysis of materials independent of their physical state. Although its great analytical potential has been proven for a broad field of applications, quantitative investigations of thin films elemental composition were reported in few studies only.

We report on the quantitative LIBS investigation of IZO compositional libraries thin films with high In/(In+Zn) concentration gradient and non-uniform thickness, synthesized by Combinatorial-Pulsed Laser Deposition. Since the optical and electrical properties of IZO thin films are strongly related to their composition, the precise determination of In/(In+Zn) ratio along the length of the samples is mandatory.

The elemental analyses were performed using an Nd:YAG laser ( $\lambda=266$  nm,  $\tau_{\text{las}}=4$  ns), in air, with a spatial resolution of  $\approx 100$ - $\mu\text{m}$ . Our investigations were based on the calculation of the spectral radiance of a plasma in local thermal equilibrium, considering Zn and In emission lines situated in the visible spectral range, taking into account the self-absorption effect. The Zn and In atomic fractions in various locations as well as the In/(In+Zn) ratio variation trend along the length of samples are assessed and compared with the results achieved by Energy Dispersive X-Ray Spectroscopy. It was found that the measurement accuracy between methods may reach values less than 5% if accurate spectroscopic data are determined in appropriate experimental conditions.

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Investigation of bulk defects formation in amorphous and polycrystalline HfO<sub>2</sub> thin film by means of temperature-resolved photo-luminescence measurements

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Even though HfO<sub>2</sub> thin films have been successfully incorporated into the gate stack of current generation MOS field-effect transistors [1], a number of open scientific and technological challenges remain relating to the properties of HfO<sub>2</sub>. One such challenge relates to electrically active defects in the bulk of the HfO<sub>2</sub> film, which exhibit a higher density of bulk defects than SiO<sub>2</sub>. These bulk defects lead to enhanced trapping in the insulator, which can influence the carrier mobility in the inversion layer, cause threshold voltage shifts, and act as a source of device instability [2,3]. As a result, there is a need to investigate the physical origin of the HfO<sub>2</sub> bulk defects and their associated energy levels, in order to select appropriate deposition and processing conditions that will minimise the concentration of defects. One approach to investigate bulk defect energy levels in HfO<sub>2</sub> films is using the low temperature photoluminescence spectroscopy.

The objective of this study is to study the formation of bulk defect in e-beam formed HfO<sub>2</sub> films by low temperature photoluminescence spectroscopy following the phase transition from the amorphous to the crystalline state. The photoluminescence spectroscopy is a well-established technique for the study of defects and it can provide direct evidence of the presence of bulk defects in HfO<sub>2</sub> films. The thermal treatment of HfO<sub>2</sub> thin films by Rapid Thermal Annealing (RTA) in N<sub>2</sub> environment from 300 up to 800°C (in 100°C steps) will allow us to follow the defect formation following the amorphous to crystalline transition while employing a consecutive thermal annealing in O<sub>2</sub> environment (1h at 600°C) it is expected to passivate the bulk defects. Based on current ab initio predictions [4] it is anticipated that most of the bulk defects are related to the formation of oxygen related defects in HfO<sub>2</sub> and therefore the extinction of the PL signal in HfO<sub>2</sub> thin films following a thermal anneal in O<sub>2</sub> it is expected to confirm the theoretical prediction,

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C. Hatzigiannakis, P. Pouli<sup>2</sup>, V. Tornari<sup>2</sup>:

## **Laser-cleaning of excavated fresco samples and DHSPI testing of model fresco samples for differentiating the photomechanical effects induced by the laser cleaning**

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Fresco fragments buried in soil are an important and frequent type of archaeological finding. Their cleaning, up to the recent times, was done mechanically, with scalpel, which method has the inherent high risk of mechanical damage. With the availability of commercial, mobile cleaning lasers, the number of publications accounting on laser cleaning of frescoes is increasing, thus the awareness of its potential negative effects is crucial.

In the ULF-FORTH001594 project, the laser cleaning of original samples (archaeological fresco findings from the Roman age) has been tested with a wide variety of experimental parameters (such as laser wavelength, pulse duration, fluence, wetting). It has been concluded that – with carefully adjusted parameters – laser cleaning can be a good alternative to mechanical scalpel-cleaning on excavated fresco fragments. It was also pointed out that photomechanical effects can play significant role in the cleaning process, depending on the characteristics of both the fresco and the contamination, as well as on the parameters of the laser pulse. The photomechanical impact caused by laser cleaning was detectable by examination of the cleaned fresco fragments with a laboratory prototype Digital Holographic Speckle Pattern Interferometry system (DHSPI system). Two interferometry criteria for assessment of impact visualisation were met: a) field alterations represented by changes in the whole interference field formation, and b) local alterations represented by limited changes of interference fringe formation. Both the field reaction of the samples and the local fringe alterations showed dynamic reaction with temporal dependence represented by relaxation in time. These results approved the importance of the investigation on the photomechanical effects of laser cleaning processes and the possible impact in laser cleaning of archaeological fresco fragments.

In the ULF-FORTH001880 project, custom made samples with encrustation of known composition were prepared in order to investigate the samples' mechanical responses to laser cleaning with the DHSPI method. The aim of the experiments was to identify the effects leading to two different types of sample responses: field reactions, that manifest in alteration of fringe shape and fringe density all over the surface, and local reactions, that result in distortions of the fringe pattern only on the affected regions. The relaxation of the samples was followed up over a month period. The features revealed by DHSPI were evaluated with respect to the different mechanical characteristics of the samples.

In our presentation, details will be given on the sensitivity of the pigmented fresco layers to the different types of lasers. Examples will be shown for satisfying cleaning results and for possible negative effects occurring due to improper experimental settings. The mechanical effects of laser cleaning will be discussed on the basis of statistical evaluation of DHSPI data acquired on model fresco fragments. The consistency between the conclusions of the DHSPI tests and the independent mechanical characterization tests will also be discussed.

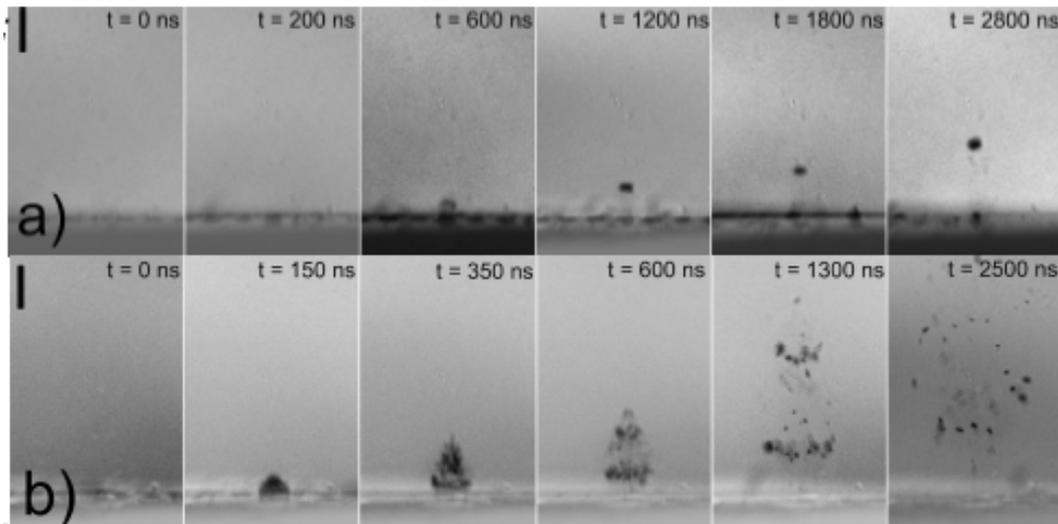
## Shadowgraph imaging in femtosecond laser-induced forward transfer of functional materials.

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Laser-induced forward transfer, (LIFT), is a laser-assisted additive direct-write method that is increasingly being used for rapid prototyping of electronic and photonic devices. Lasers with pulse durations in the ns to fs regimes are most generally used for LIFT, although fs laser sources may prove to have greater flexibility due to their much shorter pulse duration compared to ns sources, for the two important reasons of (i) multiphoton absorption within either the donor or the sacrificial release layer used and (ii) a reduced level of interfacial thermal damage.

Another important factor however is to investigate the dynamics and structural integrity of the 'flyer' (the pixel of the donor material) to be LIFT-printed, as the typical pixel size for fs lasers that may only deliver mJ level pulses, will be much smaller than those using more energetic (~100s mJ) ns laser pulses. To investigate the transfer dynamics (threshold incident laser fluence, latency and final flyer velocity) we have used the shadowgraphy set-up available at LP3, to time resolve the flyer behavior for a range of donor thicknesses and laser fluences, and typical results as shown in the figure below.



Sequence of shadowgrams showing transfer of a  $\text{Bi}_2\text{Se}_3$  flyer for a fluence of (a)  $\sim 130 \text{ mJ/cm}^2$  and (b)  $\sim 400 \text{ mJ/cm}^2$ . The times at the bottom of the frames indicate the delay times between arrival of the laser pulse and the beginning of image capture by the CCD. (The scale bar in the first frames for a delay time of 0 ns in each row is  $100 \mu\text{m}$ ).

The experiments were carried out with the help of an 800 nm Ti:sapphire laser and a flash-lamp-illuminated shadowgraph imaging system. We observed transfer of flyers in intact state at transfer velocities as low as 34 m/s for  $\sim 1.1 \mu\text{m}$  thick donors, and 48 m/s for  $\sim 1.8 \mu\text{m}$  thick lead zirconate titanate. For a  $\sim 0.5 \mu\text{m}$  thick donor of a third alloy we measured a velocity for non-intact transfer of  $\sim 140 \text{ m/s}$ . Contrary to what has been observed so far in time-resolved studies of LIFT, no shock-wave has been observed during the experiments.

## ***Session 5***

***Session Chair: Istvan Földes***

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# Excitonic absorption saturation with femtosecond UV laser pulses

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Under excitation by powerful femtosecond laser pulses, we have recently discovered an absorption saturation effect [1], which is present only for sub-band-gap excitation energies (the Urbach region). Phonon-assisted absorption, generally the accepted mechanism of absorption in the Urbach tail, cannot be applied to fs pulses, i.e. when the pulse duration is shorter or comparable to the period of optical phonons. Instead, absorption is determined by the availability of suitable atomic displacements from equilibrium positions such that the difference between the excited and ground state energies equals the excitation energy at the moment of excitation. The number of centers with an atomic configuration favorable for excitation is limited and hence leads to saturation.

The absorption saturation effect was found by studying the exciton luminescence of the CdWO<sub>4</sub> scintillator crystal using 100-fs laser pulses. Combining saturated absorption with another well-known high density effect in excitonic scintillators, the Förster energy transfer between excitons [1-4], we have developed a model which allows to study the saturation effect by analyzing the decay kinetics of excitonic luminescence. Maximum exciton densities in CdWO<sub>4</sub> (with a band gap of 5 eV) are calculated to be  $5.3 \times 10^{18} \text{ cm}^{-3}$  at 4.1 eV excitation energy with 100-fs laser pulses and  $1.9 \times 10^{19} \text{ cm}^{-3}$  at 4.66 eV excitation with 80-fs laser pulses. The approach developed is expected to be useful for similar experiments using powerful laser pulses.

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# Probing of laser - irradiated solid targets using extreme ultra – violet radiation

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## ABSTRACT

New results are presented to demonstrate that extreme ultra-violet (EUV) radiation can be employed to measure heat penetration into solid targets using the signature of a change of opacity due to heating. We examine, in particular, the effects of hot electron heating of targets by combining measurements of hot electron temperatures and fluxes in plasmas produced by irradiation of 35 fs pulses at  $3 \times 10^{16} \text{ Wcm}^{-2}$  onto targets with a preformed plasma created by a prepulse of 35 fs,  $3 \times 10^{15} \text{ Wcm}^{-2}$  incident 20 ps before the main pulse with EUV laser transmission measurements. The targets were parylene-N (CH) with a buried 50 nm thick layer of iron at a depth 50 nm below the target surface. Measured variations in the number and temperature of hot electrons with laser angle of incidence correlate with differences in the transmission to 13.9 nm EUV laser radiation, confirming modeling studies that show that the iron layer opacity changes due to hot electron heating. Such measurements of heat transport in laser-irradiated targets are important for inertial fusion studies and other applications of laser-plasmas.

# Direct Carrier-Envelope Phase Control of an Amplified Laser System

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Carrier-envelope control is one of the key technologies for attosecond pulse generation. Although the timing jitter between the carrier and the envelope has recently been demonstrated down to sub-10-attosecond range [1], CEP control is still limited to a rather narrow class of lasers, including Ti:sapphire and some other selected broadband laser materials. Typically, stabilization of a CPA laser source relies on an intricate combination of two servo loops, a fast oscillator loop and a slow amplifier loop [2]. While the second loop partially removes residual phase drift in the amplifier, unfortunately, it also corrupts the residual phase noise in the amplified pulse train due to the limited feedback bandwidth [3]. While oscillators can nowadays be stabilized down to  $\approx 10$  mrad residual CEP jitter, a 100 mrad stability for an amplified laser source is already a challenge. Here we demonstrate a direct method to stabilize the CEP of an Yb:KGW MOPA laser system, resulting in sub-100 mrad stability. To the best of our knowledge, this value surpasses most previously measured CEP jitter for Ti:sapphire CPA systems as well as any previous attempt to stabilize Yb-based oscillator and amplifier systems [4]. This is a crucial step in widening the utility of amplifier femtosecond Yb sources, which are easily scalable in average power and pulse energy but could not be meaningfully employed in attosecond optics until now, given the difficulty of their CEP control.

We extended an oscillator-type stabilization scheme behind the amplifier which resulted in a significant improvement. Our detection scheme consists of an in-line interferometer as used frequently in amplifier stabilization, yet replaces the spectrally resolved detection scheme by a simple photo-diode, as is customary in oscillator stabilization. In our experiments, we employed a commercial Yb:KGW regenerative CPA system (Light Conversion Ltd.) running at a tunable repetition rate up to 1 MHz [2]. An in-line f-2f interferometer based on white-light generation in a bulk sapphire is used for beat-note detection and out-of-loop CEP stability verification. This signal is then used to drive an acousto-optic modulator, which corrects the measured CEP in the amplified pulse train using the technique introduced in [5]. The resulting performance is depicted in Fig. 1(b-c). Using out-of-loop characterization we measure a residual phase noise of 87 mrad, see Fig. 1(b). The inset of Fig. 1(b) shows a spectral interference pattern to further illustrate the vanishing phase jitter of our scheme. Figure 1(c) shows a noise spectrum induced by the amplifier in the terms standard for characterization of oscillator stability. The significant part of the amplifier phase noise power spectrum extends beyond 50 kHz and corresponds to an  $\approx 200$  mrad integrated phase jitter. This measurement shows the necessity of the wide bandwidth phase control for the efficient suppression of the phase noise induced by the CPA laser system. We believe that our demonstration of (CEP) stabilization of an Yb:KGW MOPA laser system with record-breaking residual phase jitter below 100 mrad, opens a new perspective for high-energy CEP-stabilized parametric sources.

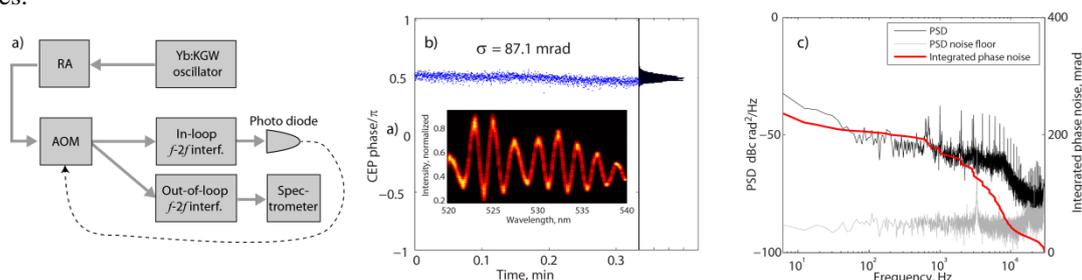


Fig 1. a) a general scheme of the frequency synthesis for the stabilization of a sub-MHz frequency comb from a regenerative amplifier (RA) b) out-of-loop CEP jitter measurement after the amplifier, inset: f-2f interferogram fringes. The spectrometer integration time was kept minimum and averaging is done over 6 laser pulses. c) measured amplifier phase noise spectrum when the oscillator CEP was locked with a feed-back loop.

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# ULTRA-FAST PULSED NEUTRON SOURCES DRIVEN BY HIGH INTENSITY LASER PULSES

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Over the past decade, significant attention has been paid to laser driven neutron sources in light of rapid developments in laser technology, and, due to possible advantages, compared to other sources, in terms of cost reduction and compactness, reduction of radioactive pollution and ability of radiation confinement by close-coupled experiments. In this context, a short burst of beamed neutron flux with narrow energy bandwidth would be particularly appealing for a wide range of applications in scientific, technological and security sectors. Specifically a narrow-energy bandwidth which is also tunable is important for improving the applicability to different probing scenarios<sup>[1,2]</sup>, as well as taking advantage of the inherent short-burst nature of the neutron source (picosecond range as compared to the  $\mu$ sec range available with conventional sources).<sup>[3]</sup>

Among the possible laser energised nuclear phenomena, the most promising route to create a neutron source is employing laser accelerated ions in either fusion or spallation reactions in a secondary target<sup>[4,5]</sup>. Employing the ELFIE laser at LULI, we explored neutron production via a beam-fusion reaction by impinging laser-driven protons accelerated via the TNSA mechanism<sup>[4]</sup> onto a secondary LiF catcher target a few mm away.

The experiment characterized, through a number of diagnostics, the neutron source produced through this reaction, as well as investigating, for the first time, neutron production with a focused, laser-driven proton beam. To this purpose, we employed a laser-triggered micro-lens technique, which can selectively focus a narrow energy band within the broadband spectrum of the accelerated protons<sup>[6]</sup>. Applying this technique allowed maintaining a high proton flux onto a LiF catcher located at a significant distance from the target, decoupling the neutron generation process from the primary interaction, which is clearly beneficial both for characterizing and applying the neutron source. In principle, this scheme has also the potential to produce neutrons from the LiF catcher, which would reduce their temporal spread after propagating to an application station, and generally enhance their capabilities in probing applications.

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**Spectral and coherence properties of the PALS X-ray laser (PALS)**

**A. Klisnick / A. Le Marec**

We present the results obtained in two experimental campaigns carried out at the PALS in March 2012 and July 2013.

Knowledge of spectral and coherence properties of X-ray lasers is essential both for fundamental reasons since they are strongly related to the plasma parameters, and for applications of these sources involving interferences or focusing (such as lithography, holography, high energy density matter generation...).

Besides, seeding X-ray lasers with high-order harmonic (HH) radiation has been recently demonstrated in optical-field ionization (OFI) and transient pumping X-ray lasers. It displayed a considerable enhancement of the coherence of the beam and is a promising way to shorten the pulse duration and increase the power of X-ray lasers. Seeding the QSS X-ray laser being currently considered at PALS, a refined experimental investigation of its spectral and coherence properties was of great interest.

Because of the extremely narrow X-ray lasers linewidth ( $\Delta\lambda/\lambda \sim 10^{-5}$ ), to measure the coherence time and characterize the spectral profile we use a specifically designed wavefront-division interferometer with variable path difference. This interferometer allows to reach extremely high spectral resolution, beyond the capability of any existing spectrometer in this wavelength range.

The first measurement of the temporal coherence of the PALS quasi-steady state (QSS) pumped X-ray laser (Ne-like Zn, emitting at 21.2 nm) has been performed in a campaign carried out in March 2012. For the first time, the related linewidth of an X-ray laser appeared to be compatible with a sub-picosecond shortest pulse duration (Fourier limit duration of the OFI and the transient X-ray lasers inferred by temporal coherence measurements  $> 1$  ps).

Another campaign has been conducted last July to refine these preliminary results. The experimental setup has been revised to increase the spatial coherence at the interferometer position and thus improve the accuracy of the measurements, and a diagnostic of the spatial coherence has been implemented.

## ***Session 6***

***Session Chair: Marc Sentis***

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## **An experiment to study Radiative Shocks relevant to astrophysics at PALS Facility**

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### **Abstract:**

Radiative shock waves are found in many astrophysical systems, including different stages of stellar evolution, but can also be created in high power laser facilities. Millimeter-scale targets are filled with a high atomic gas (xenon), and irradiated with the PALS laser, causing a shock wave to propagate in the gas at velocity  $\sim 50$  km/s. These shock waves are characterized by an induced ionization front called ‘precursor’, and their dynamics is analyzed over long time scales. The experimental results of two new diagnostics: first a XUV instantaneous imaging at 21.2 nm, and second, a time and space resolved plasma self-emission using fast diodes will be highlighted.

The work is funded by Starshock project ( ANR-08-BLAN-0263-07 - 2009 / 2013)

## Investigation of energy transfer from iodine laser shock wave generated in solid target relevant to shock ignition

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Efficiency of laser radiation energy transport into shock wave generated in layered planar targets (consisting of massive Cu coated by thin CH layer) was investigated. The targets were irradiated using two laser pulses. A  $1\omega$ -pulse with the energy of  $\sim 50$  J produced a pre-plasma imitating the corona of the pre-compressed ICF target. Second main pulse used the  $1\omega$  or  $3\omega$  laser harmonics with the energy of  $\sim 200$  J. The influence of the pre-plasma on parameters of the shock wave was determined from the crater volume measurements and from the electron density distribution measured by 3-frame interferometry. Experimental results show that the energy transport by fast electrons provides a definite contribution to the dynamics of the ablative process, to the shock wave generation and to the ablation pressure in dependence on the target irradiation conditions. The strong influence of the pre-plasma on the investigated process was observed in the  $1\omega$  case .

The experimental results together with theoretical analysis support will be presented.

# Nucleus-electronic cloud coupling in plasma: the case of $^{84}\text{Rb}$ .

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The development of high-intensity lasers is opening up new opportunities for nuclear-physics studies. Lasers are promising tools to study nuclear properties in extreme plasma conditions, which cannot be reached with conventional particle accelerators. In a plasma, the interaction between a nucleus and its electronic cloud can be influenced by its environment and unusual excitation processes such as nuclear excitation by electronic capture (NEEC) and nuclear excitation by electronic transition (NEET) should be observed. Indeed, NEEC (unobserved) and NEET (observed in  $^{197}\text{Au}$  [1],  $^{189}\text{Os}$  [2,3] and  $^{193}\text{Ir}$  [4] in accelerator based experiments) are exotic processes relevant to astrophysics, which can be dominant in particular plasma conditions of temperature and density [5], and therefore it is highly desirable to study them. These two processes involve a coupling between the nucleus and the electronic cloud. In the case of the NEET, a bound-bound atomic transition leads to a nuclear excitation if the two transitions are resonant and have the same multipolarity. The NEEC process is equivalent to the NEET process with the difference that the atomic transition is bound-free. We have undertaken a joint experimental and theoretical program to investigate the  $^{84\text{m}}\text{Rb}$  excitation rate in laser produced plasma.

The long-lived isomeric state of the  $^{84}\text{Rb}$  (energy of 463.6 keV,  $T_{1/2}=20.26\text{min}$ ) can be excited towards a higher lying short-lived state. According to the literature, the nuclear transition energy involved in this excitation is 3.05 (20) keV. It is possible to find atomic transitions, which match this nuclear transition for charge states between  $27^+$  and  $32^+$  allowing the NEET process to take place [6]. Searching for the NEET process in plasma requires an accelerator to produce the  $^{84}\text{Rb}$  isomeric state and a high-energy- high-intensity laser to create the plasma with the required charge states. This kind of facility exists at GSI (Darmstadt, Germany) where the PHELIX high-energy laser is combined with the UNILAC ion accelerator.

To prepare the NEET experiment, nuclear and atomic physics experiments were conducted. We have made experiments at ELSA (CEA/DAM, Bruyères-le-Châtel) and at TANDEM/ALTO (Orsay) accelerators to measure the nuclear transition energy with high accuracy. Indeed, the accuracy of this transition energy in the tables was insufficient (200 eV) in comparison with the accuracy of the atomic data (few eV) [7]. We have also measured the X-ray spectra emitted by a Rb plasma at PHELIX. These spectra will be compared with theoretical ones to determine plasma conditions (charge states, temperature...) reached during the experiment. If these conditions are favorable for the observation of the NEET effect, the final NEET experiment will be conducted. The results of these preparation experiments will be presented.

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# Diffuse optical tomography with fast-gated SPADs

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We present experimental results of time-resolved reflectance diffuse optical tomography (DOT) performed with fast-gated single-photon avalanche diodes (SPADs) and show an increased imaged depth range for a given acquisition time compared to the non gated mode.

In the context of analyzing biological tissue with near-infrared light, the possibility to realize optical probes with small interfiber distances to study deep layers with reflectance measurements is currently investigated. This specific geometry is relevant to probe organs whose access is anatomically restricted like the prostate. But for organs like the breast or the brain it can also be preferred to other optodes configurations for other reasons: small probes are practical to handle for the medical practitioner and easy to position on the patient compared to rings of fibers for which a wrong placement causes errors on the obtained results. In the case of reflectance measurements at small interfiber distances, time-resolved techniques enable to reach high sensitivity to deep layers of the probed tissue by selecting late photons. We have proposed an image reconstruction method for DOT based on the Mellin-Laplace transform (MLT) of Time-Point Spread Functions (TPSFs) which allows including the information of late photons in the reconstruction process [1]. We have shown that this method can be adapted to the dynamic range of the TPSF, and enables to detect and localize deeper absorbing inclusions in a diffusing medium for a higher dynamic range [2].

Recent developments with fast-gated single-photon avalanche diodes (SPADs) associated to a time-correlated single-photon counting (TCSPC) setup have shown that it is possible to acquire TPSFs with a high dynamic range in limited acquisition time compared to classical non gated mode [3]. This technique has already been applied to turbid media for improving the contrast to deep absorbing inclusions in reflectance.

In this study, we show that a setup involving fast-gated SPADs (Figure 1) can be used to reconstruct images of the absorption coefficient in turbid media with DOT algorithms (Figure 2). In particular we demonstrate that the reconstructed depth range and depth localization accuracy of a single absorbing inclusion in a turbid medium are improved by using the fast-gated mode, for an interfiber distance of 15 mm.

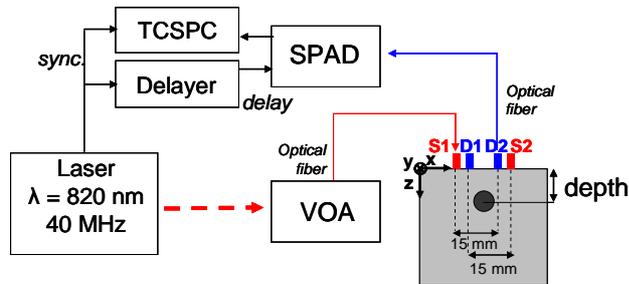


Figure 1. Experimental setup and view of the probe and phantom (2D cross section in plane  $(x, z)$ ). The SPAD module is described in [4]. The probe contains two couples of source and detector S1D2 and S2D1, both having the interfiber distance of 15 mm. Background medium:  $\mu_a = 0.1 \text{ cm}^{-1}$ ,  $\mu'_s = 10 \text{ cm}^{-1}$ . Absorbing inclusion:  $\mu_a = 0.8 \text{ cm}^{-1}$ ,  $\mu'_s = 10 \text{ cm}^{-1}$ .

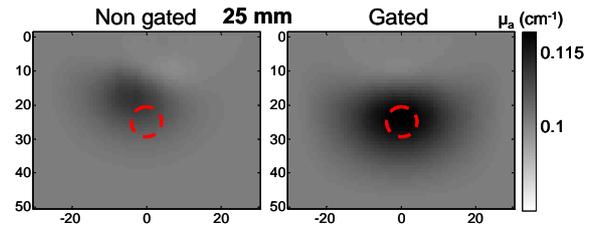


Figure 2. Reconstruction results: maps of the absorption coefficient in plane  $(x, z)$  for an absorbing inclusion at the depth of 25 mm, when using the SPAD in the gated mode and not in the gated mode (red circle: real inclusion,  $x$  and  $z$  axis in mm). Using the gated mode enables to localize precisely the absorbing inclusion at the depth of 25 mm.

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# Characterization of hot electrons in dense plasmas by high-resolution X-ray imaging near 8 keV

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The characterization of suprathermal electrons is a key subject for a large scientific community, particular in fusion science, high-intensity laser-matter interaction, atomic physics and diagnostics:

- a) in the indirect drive inertial fusion scheme, suprathermal electrons (either in the hohlraum itself or inside the capsule) are a major topic in the ignition campaign,
- b) in the direct drive (fast igniter) suprathermal electron fraction are supposed to deposit energy to a pre-compressed capsule to initiate ignition (note that propagation of high current densities exceeding  $10^{14}$  A/cm<sup>2</sup> request return currents),
- c) in dense high current pinching plasmas, the generation of suprathermal electrons is strongly connected with the onset of turbulence and the explosion of the pinched plasma,
- d) the study of the radiative properties is an important subject in atomic physics that invokes non-Maxwellian atomic population kinetics (challenging as hot electrons provoke important core-hole ionization and associated satellite transitions) [1].

The development of non-Maxwellian and time dependent atomic physics models and spectroscopic diagnostics is therefore an important activity to provide independent (from plasma simulations) information on suprathermal electrons.

We report here on a recent experimental campaign [2] carried out at the PALS laser facility to study hot electron signatures via high-resolution X-ray spectroscopy of the target emission. Irradiating copper targets at 1 and 3  $\omega$  with several 100 Joule laser energy and employing curved quartz Bragg crystals allowed to measure significant qualitative changes in the near 8 keV X-ray spectral distribution. Apart the possibility to characterize hot electron fractions and directions we will discuss the relevance of ns-installations to impact on future developments of non-LTE & non-Maxwellian atomic physics codes.

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- [2] LASERLAB-project: "*Exploration of high-resolution K-shell X-ray emission of Cu and Ge to study suprathermal electrons in dense hot matter*", experimental campaign 04-05/2013.

# High resolution x-ray spectroscopy of laser-produced solid-density plasmas

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Studies of warm dense matter (WDM) are an emerging and challenging field that is at the crossroads of condensed matter physics and plasma physics. The WDM state nowadays is routinely formed in laboratory in the interactions of intense laser or particle beams with solid-density matter. During this interaction electrons are accelerated to relativistic energies. This is accompanied by the creation of strong electric fields, reaching  $\sim$ TV/m. Because of the high density, only short wavelength radiation may escape the inner parts of the target. Due to this reason, the inner-shell x-ray emission is an important instrument to yield valuable information about such plasmas. Formerly, the temperature and K-yield radial distributions in laser-produced solid-density plasmas were measured using the 1D spatial imaging x-ray spectroscopy [1]. However, the integration along the z axis results in a weighted averaging of the spectra observed by accumulating photons emitted from different depths of the target. In other studies, sandwich targets were used to achieve axial resolution, although new complications arise due to the layer bonding and contact effects. Instead, a novel non-intrusive technique, based on 1D-imaged spectrally-resolved multi-angle measurements was employed in order to reconstruct the 3D K-line emissivity. Also, an experimental setup aimed for measuring the strong electric fields created at the surface of the target will be discussed [2] and preliminary results will be presented.

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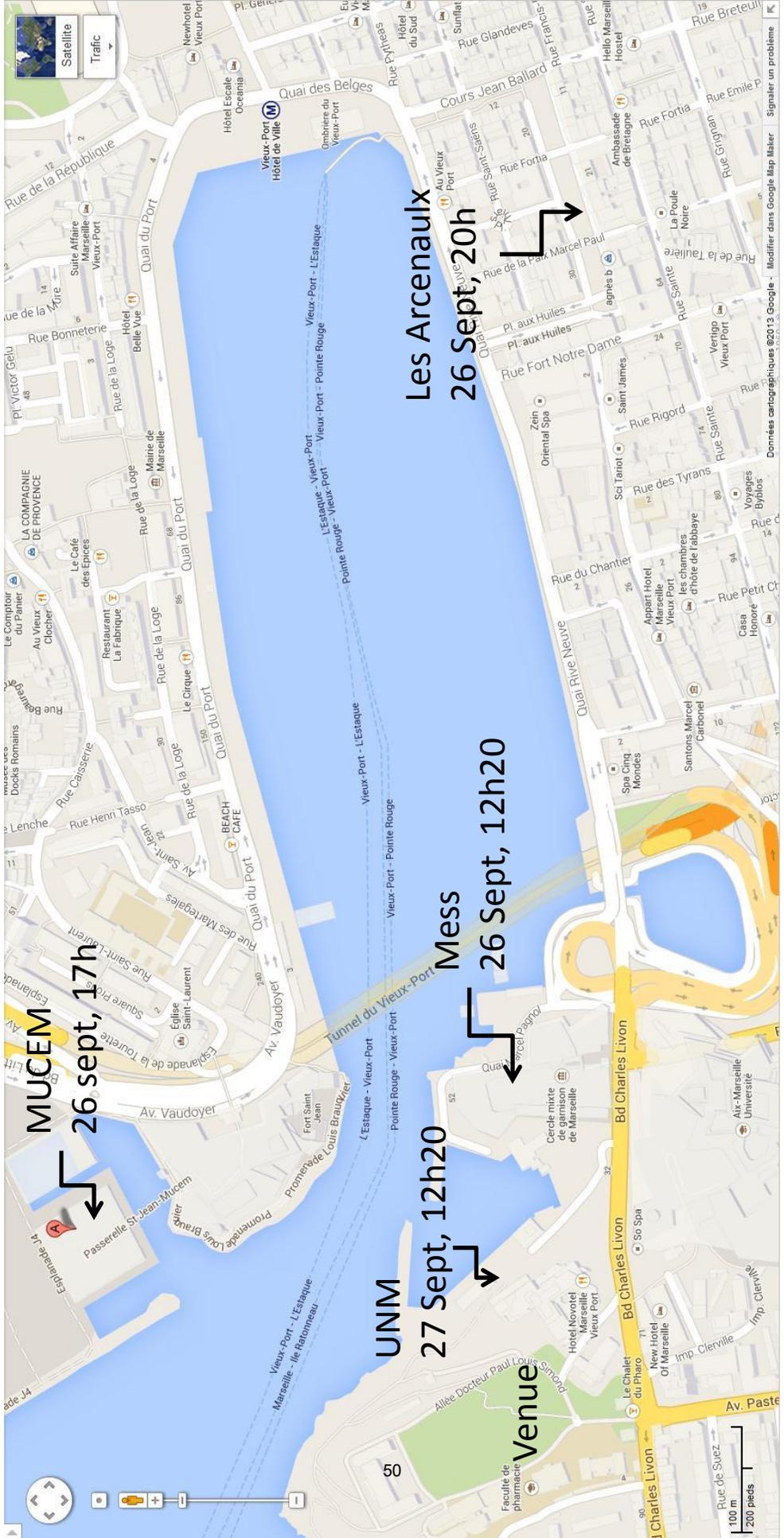
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Quantum Electronics Department and Laser Research Center, Vilnius University, Vilnius, Lithuania

# *Maps*

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**MUCEM**

26 sept, 17h

**UNM**

27 Sept, 12h20

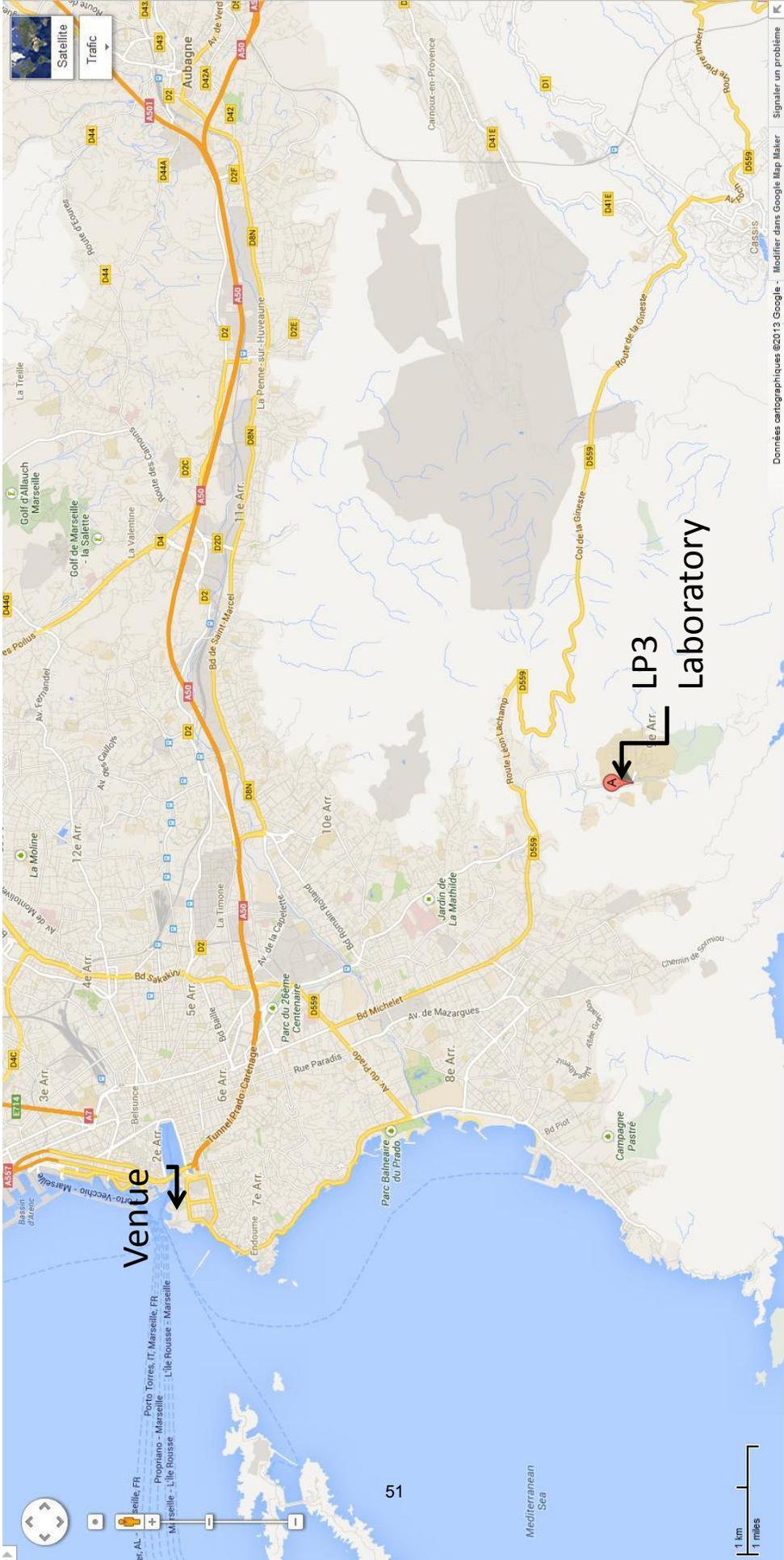
**Mess**

26 Sept, 12h20

**Les Arcenaux**

26 Sept, 20h

100 m  
200 pieds



Venue

LP3

Laboratory

## *Notes*

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