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Laserlab Europe

Laserlab Forum

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the integrated initiative of European laser infrastructures funded by the European Union's Horizon 2020 research and innovation programme

Lasers and Clean Energy

Burning iron particles
Photo: Jianqing Huang/LLC

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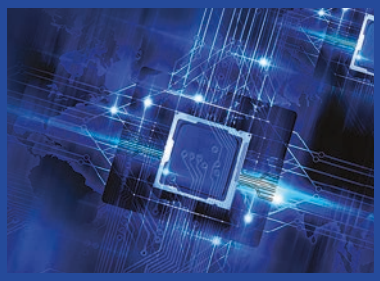

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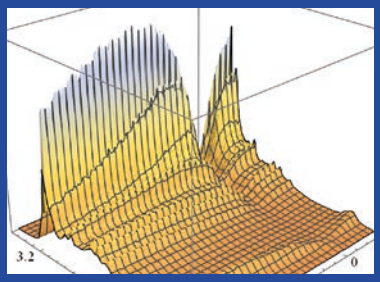

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Editorial

Sylvie Jacquemot

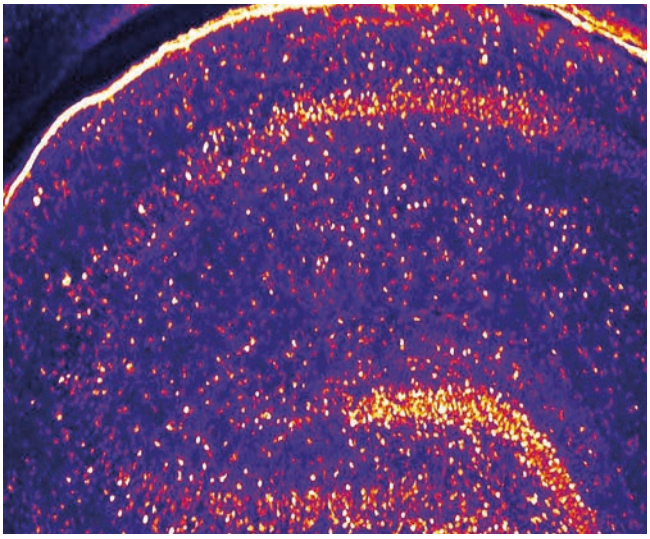
The Covid-19 outbreak has had a significant impact on Laserlab-Europe and its partners. Most laser research infrastructures had to fully or partly close down during the first phase of the crisis, inducing a strong limitation in their networking and transnational access activities; many users were thus asked to cancel or postpone their experiments. However, where possible, remote fast-track access for COVID-related research was implemented early on. Since then, safety measures have been put in place and the facilities have changed their operating practices allowing many to reopen and host external users. In addition to on-site hands-on experiments, it is also now possible to conduct transnational access projects remotely at a large majority of the Laserlab-Europe facilities. It is one of the initiatives launched to maintain, and even enlarge, our service to users during the coronavirus crisis. Coping with difficulties may open up new opportunities; like the scientific community as a whole, Laserlab-Europe is jointly developing alternative plans and formats to meet its needs, not only making the best out of the current situation but also with a view to integrating the advantages of new solutions into its future operations.

The present newsletter includes examples of such new operations while the focus of the scientific highlights addresses a major societal challenge: clean energy. Have a pleasant reading of this first 2021 issue of the Laserlab-Europe newsletter!

Finally, I take the opportunity of this editorial to wish you the very best in the coming year.

Sylvie Jacquemot

News



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2-photon image of a mouse brain slice with GFP labelled neurons.

Next-generation microscope to revolutionise the study of the cellular origin of diseases

CRIMSON, a trans-disciplinary and transnational research project recently granted 5 million euros over 42 months by the European Commission, aims to revolutionise precision medicine by developing the next generation of bio-photonics imaging devices for biomedical research, combining advanced laser techniques with artificial-intelligence data analysis. It can provide 3-D quantitative maps of sub-cellular compartments in living cells and organoids and enables fast tissue classification with unprecedented biomolecular sensitivity. High acquisition speeds will allow the observation of intra and inter-cellular dynamic changes by time-lapse imaging.

The consortium is coordinated by Politecnico di Milano (Italy) and features three research centres (POLIMI, Leibniz IPHT, Germany, and Institut Fresnel, CNRS, France), who will use their long-standing expertise in photonics, spectroscopy and nonlinear microscopy to develop the technology. Three biomedical partners (Istituto Nazionale Tumori, Institut National de la Santé et de la Recherche Médicale, and Jena University Hospital) will validate the imaging system on open biological questions. More info is available at www.crimson-project.eu

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The partially-completed new building at CALT.

National Calibration Laboratory for Time and Frequency approved

Within the framework of the CALT project, funds have been provided for the equipment of the National Calibration Laboratory for Time and Frequency of the Republic of Croatia. The new laboratory will provide traceability to the SI standards for time and frequency, and will measure and disseminate Croatian UTC time. In addition, the laboratory will measure high-stability frequency in the radiofrequency range and develop, for the first time in south-eastern Europe, a quantum clock based on cold strontium atoms. Strontium's atomic clock is the best atomic clock of our time in terms of complexity and stability, and also a candidate for redefining the second.

In addition, the new laboratory will participate in determining the world timescale and the atomic weight scale, and is aiming for accreditation and active participation in the International Bureau of Weights and Measures (BIPM), the European Association of National Metrology Institutes (EURAMET), and to have its capabilities listed in the International Database (KCDB).

Przemysław Wachulak new Rector of Military University of Technology

On 31 August 2020, on-site at the MUT, the official ceremony to inaugurate a new Rector took place. In the presence of Secretary of State Wojciech Skurkiewicz, the responsibilities of Rector of MUT were passed from Prof Tadeusz Szczurek to Prof Przemysław Wachulak, long-time researcher at the Institute for Optoelectronics.

The highlight of the ceremony was the symbolic handing over of the MUT banner. The Secretary of State offered special acknowledgement to the former Rector for his service, commitment and work, emphasising that MUT is now a leading-edge institution in Poland. The new Rector remarked that the unique importance of MUT is its long-term scientific and teaching achievements, and that MUT carries out research at the highest world level, adding that he will do his utmost to bring the university to the heights of its scientific and didactic development.

What is Laserlab-Europe?

Laserlab-Europe, the Integrated Initiative of European Laser Research Infrastructures, understands itself as the central place in Europe where new developments in laser research take place in a flexible and co-ordinated fashion beyond the potential of a national scale. The Consortium currently brings together 35 leading organisations in laser-based inter-disciplinary research from 18 countries. Additional partners and countries join in the activities through the association Laserlab-Europe AISBL. Its main objectives are to maintain a sustainable inter-disciplinary network of European national laboratories; to strengthen the European leading role in laser research through Joint Research Activities; and to offer access to state-of-the-art laser research facilities to researchers from all fields of science and from any laboratory in order to perform world-class research.

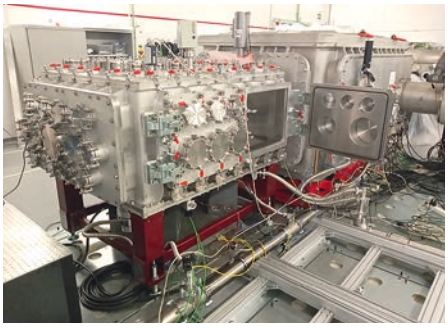
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Image of the new Petawatt chamber at VEGA.

New investment for VEGA

The VEGA system at CLPU has received an investment grant from the EU Regional development fund of € 287,300 for the supply and installation of new equipment. This will permit the reorganisation and enhancement of the "PHASE I" pump-probe capabilities at VEGA, for example by installing a new vacuum chamber in the Petawatt line, which is more flexibly designed with arrays of ports properly oriented to couple to the various diagnostics, as well as new diagnostic devices such as a Streak Camera with a few-ps temporal resolution and a new ultra-low-noise X-ray CCD which can detect between 10 eV and 30 keV.

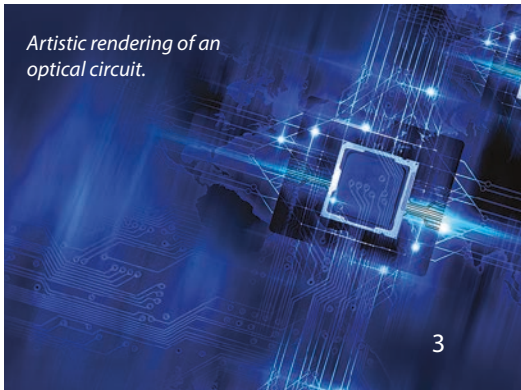
Analytical Research Infrastructures network founded

Earlier this year, seven European networks have joined forces in the network of Analytical Research Infrastructures in Europe (ARIE), consisting of Laserlab-Europe (lasers), LEAPS (photons), RADIATE (ions), INSPIRE (protons), EMFL (high magnetic fields), LENS (neutrons), and e-DREAM (electrons). In July, ARIE released a set of position papers highlighting how their complementary approach will help address the societal challenges Europe is facing and which are the focus of the Missions in the Horizon Europe framework programme, starting in 2021. The papers give input for the strategic planning of the Missions and outline the kind of advanced experiments and research that the ARIE facilities can perform in support of the Missions.

To address the Missions, the transversal platforms of ARIE will collaborate amongst themselves and with the Mission specialists at unprecedented levels. They will build and exploit open networks to share knowledge and skills, to coordinate access, to prepare samples, and to create the sample environments required for experiments under real conditions, making use of EU-wide programmes such as the new European Open Science Cloud.

OPTOlogic to develop faster, more energy-efficient electronic circuits

Topologically protected electronic states can exhibit the amazing phenomena of robust, lossless electronic transport. However, realising devices based on topological materials has proven difficult. The EU-funded project OPTOlogic aims to take a novel approach to address this, by creating and engineering light-induced topological states in 2D materials through the use of precisely tailored, ultrafast laser pulses. Coordinated by ICREA Prof Jens Biegert at ICFO, the OPTOlogic consortium includes researchers at ICFO, the Fritz Haber Institute of the Max Plank Society, the French Alternative Energies and Atomic Energy Commission (CEA) at Saclay, the Max Born Institute Berlin, and the company LightOn.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Artistic rendering of an optical circuit.

ERC Synergy Grant

ERC Synergy Grants fund research projects with groups of two to four outstanding researchers, who have complementary skills, knowledge and resources. The intention is to enable scientists to address ambitious research questions using an innovative multidisciplinary approach. 34 projects were funded out of more than 440 submitted proposals in this Synergy Grant call, making it one of the most competitive calls according to the ERC.

Fernando Martín García

Mauro Nisoli

Nazario Martín León

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The European Research Council (ERC) has announced that the project TOMATTO – "The ultimate time scale in Organic Molecular opto-electronics, the ATTOsecond", led by Fernando Martín (IMDEA Nanoscience and Universidad Autonoma de Madrid), Mauro Nisoli (Politecnico di Milano) and Nazario Martín (Universidad Complutense de Madrid) has been awarded a Synergy Grant of almost 12 million euros for the next 6 years.

The ultra-fast motion of electrons which is induced by interaction with light is the basis of the conversion of solar energy into electrical energy. It plays a crucial role in fundamental processes in nature such as photosynthesis, the transport of signals in biological molecules, the mechanisms of DNA damage, and many others. The common denominator in all these processes is the absorption of light and the generation of microscopic electric currents. What happens inside the individual molecules immediately after the interaction with light is still a mystery, because light initiates events that evolve on extremely short time scales, of the order of attoseconds.

TOMATTO aims to film the motion of electrons induced by light in molecules, with an unprecedented temporal resolution. The ultimate goal is to design molecular materials with improved opto-electronic properties. To do this, a new attosecond laboratory at the forefront of the international arena will be built at the Attosecond Research Centre of the Politecnico di Milano, a new supercomputer incorporating the latest advancements in hardware and software developments will be installed at the Computer Centre of the Universidad Autonoma de Madrid, and new opto-electronic organic materials, with still unforeseen capabilities, will be synthesised at Complutense University Madrid. Work will be led by a team of experts in laser technologies, the synthesis of new organic materials, and computational methods.

The project synergistically combines the development of innovative laser measurement methods with extreme time resolution, with the most advanced methods of organic synthesis and computational modelling. The principle investigators said: "Understanding how light interacts with matter on the attosecond time scale and how the ultra-rapid motion of electrons depends on the molecular structure, are in themselves extremely important scientific objectives. The ability to understand and control these processes on the time scale of attoseconds offers the possibility of opening up new research fields beyond the scope of the project. In particular, we foresee important applications to the study of light-guided processes in a variety of both natural and artificial structures, ranging from systems of biological interest, to advanced materials with new functionalities. TOMATTO has the potential to lead to important and not easily predictable discoveries and advances: a typical example of high-risk high-gain research."

Lasers and Clean Energy

Clean energy is a very broad field of research, ranging from fundamental science (such as charge recombination dynamics in artificial photosynthesis) and specific technologies (such as improving light accumulation on a solar panel or reducing particulate emissions in a particular process). At Laserlab-Europe, research is being done across this spectrum, bringing clean energy closer to reality.

Can advanced laser diagnostics help to develop metal fuels as a replacement to fossil fuels? (LLC, Sweden)

One of the most important things for mankind is a safe, reliable and sustainable energy supply. At the moment, ≈ 80 % of the worldwide energy utilisation comes from combustion, where fossil fuels, e.g. coal, gasoline, diesel, are in the majority. Clearly, due to the limited resources and environmental impact, mainly from the large emission of carbon-dioxide, these fossil fuels have to be phased out in the near future.

An alternative fuel, which has seen little consideration thus far, is the use of metal powders, e.g. iron, aluminium or silicon. These fuels have the great advantages that the energy density is higher than those of fossil fuels, the combustion takes place without any emission of carbon-dioxide, polycyclic aromatic hydrocarbons (PAH) or soot, and furthermore the fuel is transportable and relatively safe to store. Clearly, as a new fuel there are many aspects that are unknown and which have to be investigated in detail in the laboratory before this fuel can be used by society to replace fossil fuels.

© Jianqing Huang

Burning iron particles.

As part of this necessary development, the use of advanced laser diagnostic techniques can be instrumental. These techniques can provide non-intrusive measurements with high spatial and temporal resolution. The parameters that are of primary importance are species concentration, temperature, flow velocity and particle characteristics.

The Division of Combustion Physics within Lund Laser Centre (LLC) has for many years been developing and applying different laser diagnostic techniques for characterising various combustion phenomena. Thanks to recent financial support, it has also been possible to develop and apply laser techniques for further development and understanding of metal combustion. When using laser diagnostics in a particle environment, special attention has to be paid to the strong Mie scattering as well as multiple scattering which may spectrally interfere. However, similar challenges are seen when working in conventional fuels, e.g. in spray combustion. A technique based on structured illumination can be used here: structured laser illumination planar imaging, whereby the multiple scattering can be suppressed. Furthermore, a new development of conventional Raman scattering based on periodic shadowing, can be applied. When using this technique, a development to suppress background based on lock-in of polarisation rotation as well as time gating can also be utilised. Another approach is to use various coherent techniques, e.g. coherent anti-Stokes Raman, degenerate four-wave mixing or laser-induced grating spectroscopy. Here, the signal is generated as a new laser beam and all particle scattering can be simply suppressed using apertures.

So to summarise, by using appropriate laser diagnostics and in close collaboration with modelling expertise in computational fluid dynamics and chemical kinetics and with industry, the aim and hope is to develop these new and fascinating fuels to the benefit of our society and for our environment.

Marcus Aldén (LLC)

Enhanced hydrogen production through electrolysis by novel, laser-induced nanostructured nickel electrodes (ULF-FORTH, Greece)

Researchers from the Ultraviolet Laser Facility at FORTH (ULF-FORTH) in collaboration with the Laboratory of Matter Structure and Laser Physics at the Technical University of Crete (LMSLP-TUC) have initiated experimental research using ultrashort laser pulses to fabricate 2D and 3D nanostructures for applications in sustainable, carbon-free and eco-friendly energy production and storage. An important milestone towards this goal is to produce nanostructured electrodes for use in hydrogen production through electrolysis or for energy storage in batteries.

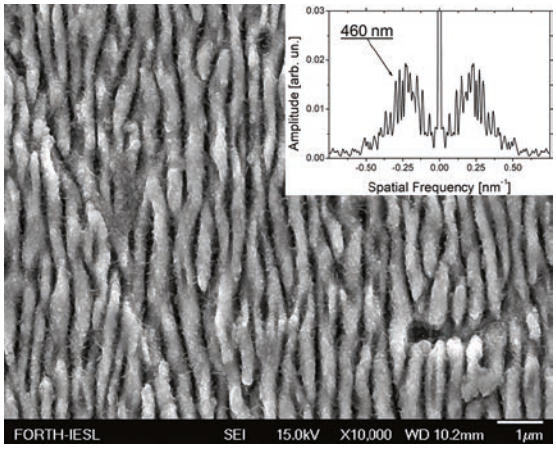


Figure 1: SEM image of the laser-nanostructured nickel surface. Inset shows the Fourier transform of the SEM image.

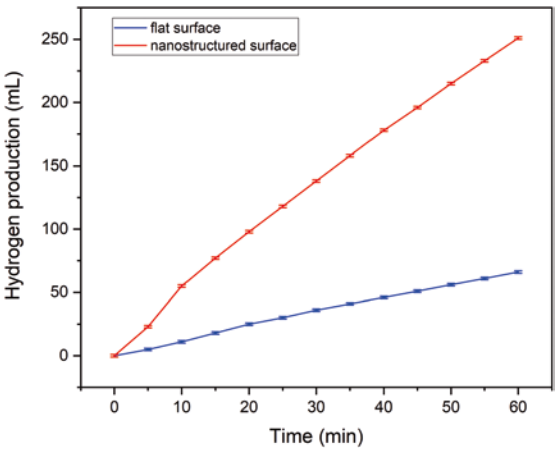


Figure 2: Hydrogen gas production comparison between an untreated nickel electrode and a laser-nanostructured electrode, for one hour of electrolysis.

Nickel is among the top materials to be used as the template to produce the laser-nanostructured electrodes for micro-batteries. In this work, laser structured nickel electrodes are produced and tested for the production of hydrogen gas in alkaline electrolysis. The nickel surface was laser-scanned using optimal conditions (fluence, pulse number per spot) for production of laser-induced periodic surface structures (LIPSS, or "ripples"). LIPSS are known to be the result of the interaction of the incident laser field with a surface wave, created by the laser itself (the surface plasmon). The spacing of the lines produced during the scanning of the metallic surface with the laser beam was chosen to ensure that the metallic surface was densely covered by the ripple nanostructures, i.e. slightly overlapping scan lines. A scanning electron micrograph (SEM) of the laser-scanned metallic surface is shown in Figure 1. Almost homogeneous ripples are shown to cover an extended metallic surface. The ripples form a periodic pattern with a period that is dictated by the parameters of the incident laser field (wavelength, intensity). By performing a Fourier transform on the SEM image, shown at the inset of Figure 1, the period of the nanostructured ripples was found to be 460 nm with a relatively narrow distribution.

A custom-made electrolysis cell was set up in order to evaluate the performance of the nanostructured electrodes in the production of hydrogen, during the hydrogen evolution reaction (HER). The electrolyte was 1 Mole potassium hydroxide solution, the anode was untreated nickel and the potential was kept steady at 3 V. It was found that the densely nanostructured electrode produces 3.7 times more hydrogen than an untreated (flat) one (Figure 2). This increase is due to the creation of periodic nanostructures and additional nanoparticles on the electrode surface created by the laser interaction. These increase the electrochemical active surface area, which in turn leads to an increase at the production rate of hydrogen gas.

This approach has demonstrated that laser structured nickel electrodes do produce larger quantities of hydrogen gas, and that they can be used not only as hydrogen storage electrodes but also as cathode electrodes in alkaline electrolysis HER. This approach may help the long or short scale timeline of hydrogen production, because of its low cost, short time of manufacturing and the large quantities of produced hydrogen. Further work is underway in order to optimise the electrode characteristics for optimally efficient hydrogen production or for use in energy storage in batteries.

Panagiotis Loukakos, Maria Farsari, Michail Tsanakas, Nicandra Papakosta, Argyro Klini (FORTH), Ioannis Poimenidis, Stavros Moustaizis (TUC)

Ultrafast optical spectroscopy sheds light on the primary steps of solar fuel production (CUSBO, Italy)

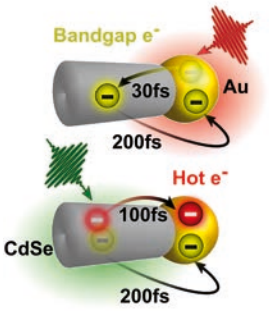
Within the European Green Deal, a key challenge is to decarbonise the energy sector and provide renewable, clean energy. Sunlight is a potential long-term solution: ≈ 0.05 % of incident solar energy would meet current global needs. However, harvesting this energy must be both efficient and cost-effective.

For the two approaches to solar harvesting (photovoltaic and photocatalytic), a key aspect is the development of novel materials with higher efficiencies or simpler production processes. For photocatalysis, the focus is on nanostructured materials, in which light absorption is followed by charge separation across an interface. This is then used to drive a redox reaction, e.g. reducing H_20 into hydrogen. Ultrafast optical spectroscopy plays a key role in these research efforts, as it illuminates both the mechanics of photoexcitation in molecules/materials and the real-time light-to-energy conversion process.

Semiconductor-metal hybrid nanoparticles (HNPs), in which a semiconductor nanostructure is coupled to a noble-metal co-catalyst, offer a highly controllable platform for light-induced charge separation and photocatalysis [1]. The many degrees of freedom (shape, size and composition of both semiconducting and metal components) allow tunable electronic and optical properties, but also require careful optimisation.

Prof. Uri Banin's group (Hebrew University of Jerusalem), together with the team at the Centre for Ultrafast Science and Biomedical Optics (CUSBO), Politecnico di Milano, used ultrafast spectroscopic tools to decipher the complex charge transfer pathways in HNPs consisting of semiconductor nanorods with gold tips (see figure below). First, the CdSe-Au HNP model system was used to study the effect of the tip size on photocatalytic function. Combining transient absorption, hydrogen evolution kinetics, and theoretical modelling revealed a non-monotonic behaviour with the size of the gold tip [2]. Considering the size-dependent interplay of the metal domain charging, the relative band-alignments, and the resulting kinetics, an optimal tip size was found. Using nonlinear excitation, in which multiple excitons are generated in the semiconducting component, revealed a competition between Auger recombination and charge separation: the dominant process switched from Auger recombination to charge separation with increasing size of the metal domain, allowing for effective multiexciton dissociation and harvesting in large-metal-domain HNPs, and an improvement of their photocatalytic activity [3].

Very recently, CdSe-Au HNPs was studied because it manifests strong hybridisation of the excitonic and plasmonic states. In these systems, charge separation can occur following photoexcitation of either the semicon-



Ultrafast optical spectroscopy is used to visualise in real time the forward and backward electron transfer processes in metal-semiconductor hybrid nanoparticles, which occur on the 100-fs timescale.

ductor or the localised surface plasmon resonance (LSPR) of the metal. In particular, a novel mechanism of plasmon induced charge transfer (PICT) was discovered, whereby the LSPR excitation is directly accompanied by a rapid charge separation, creating an electron in the semiconductor and a hole in the metal. By applying spectroscopy with >10-fs resolution, the complete pathway of electron transfer was resolved for both semiconductor and LSPR excitation. The LSPR experiences an ultrafast (< 30 fs) electron transfer to CdSe via PICT, followed by back transfer from the semiconductor to the metal within ≈ 200 fs (see figure above).

These studies show the power of ultrafast optical spectroscopy to identify the complex nanoscale photophysical processes involved in solar energy conversion in HNPs and to measure the corresponding timescales. These results establish the design parameters for HNPs in a photocatalytic material.

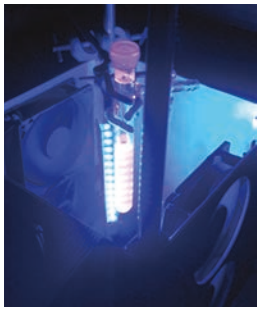
Giulio Cerullo, Francesco Scotognella, Sandro De Silvestri (CUSBO) and Uri Banin (Hebrew University Jerusalem)

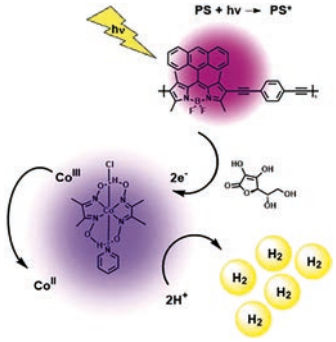
[1] P. Kamat, J. Phys. Chem. Lett. 3: 663–672, 2012
[2] Y. Ben-Shahar et al., Nat. Commun. 7: 1–7, 2016
[3] Y. Ben-Shahar et al., Nano Lett. 18: 5211-5216, 2018

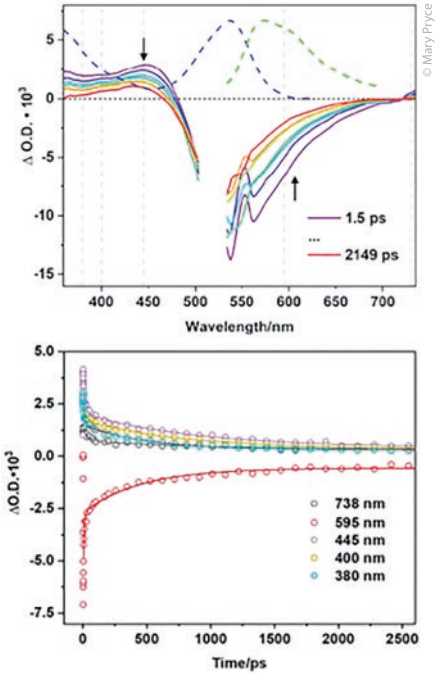
Using time-resolved spectroscopy to identify the excited states leading towards hydrogen generation (CLF, UK)

New approaches to the generation of sustainable energy sources are high on political agendas. This is for a range of reasons, including climate change, depleting fossil fuels, and a predicated increase in energy requirements. Currently, the main source of energy is fossil fuels, which when burned release pollutants and most importantly greenhouse gases. An alternative approach for energy generation is the use of sunlight. This can be harnessed by using photosensitisers to split water into hydrogen and oxygen.

An aim of the research of Dr Mary Pryce (Dublin City University, Ireland) in collaboration with Dr Elizabeth Gibson (Newcastle University) is to devise new, robust, cheap, and abundant photocatalytic materials that are capable of splitting water into hydrogen and oxygen. In this way, solar energy can be effectively stored as a solar fuel (H_2). Time-resolved techniques, such as time-resolved absorption spectroscopy (TA) and time-resolved infra-red spectroscopies (TRIR), are essential tools in characterising the excited states leading to hydrogen generation from water. For example, in a recent Laserlab-Europe access project (EU access grant 16140001 and CLF002565) with the UK Central Laser Facility (CLF), they used both TRIR and TA to identify the excited states generated in BODIPY copolymers with conjugated co-monomers [1][2]. They used these polymers to generate hydrogen from water in the presence of a cobalt catalyst. The time-resolved techniques provided important insights into the reactivities of reactive intermediates in the hydrogen generation process. Armed with this data, it is possible to design and develop superior photocatalytic materials, to enhance the efficiency of hydrogen generation.







© Mary Pryce

Using time-resolved spectroscopy to identify the excited states leading towards hydrogen generation.

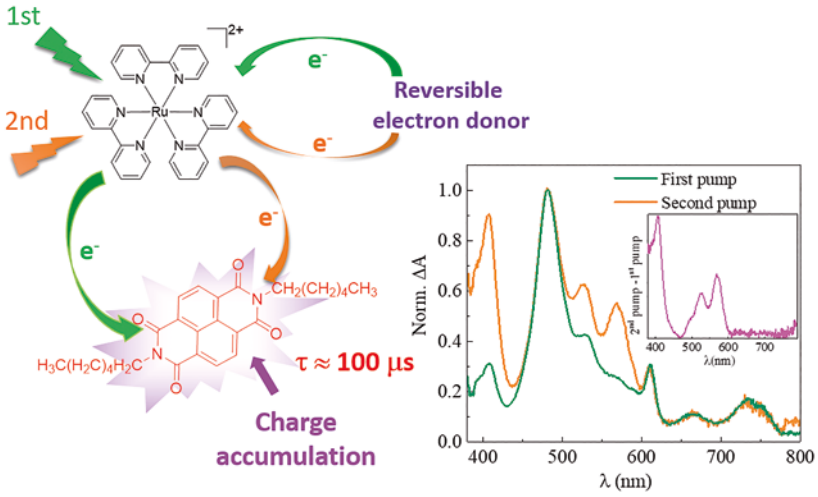
In addition to this water-splitting research, UK-based research groups have used the facilities at the CLF to apply time-resolved techniques to track the reaction pathways in Earth-abundant organometallic [3] and organic [4] homogeneous catalysts. For this both TRIR and pump-IR probe-probe-probe (TRMPS) vibrational spectroscopy have proven invaluable in tracking the reaction pathways of model systems all the way from picosecond to millisecond timescales. The knowledge this provides of the steps involved in the catalytic cycles will allow rational approaches to the optimisation of catalysts. The new mechanistic understanding gained helps in the aim of developing Earth-abundant metals as competitive replacements for precious metal catalysts, as well as driving towards cleaner and cheaper means of forming industrial chemical precursors and complex molecular architectures.

Mary Pryce (DCU) and Mike Towrie (CLF)

[1] A. A. Cullen et al., Front. Chem. 8: 957, 2020
[2] N. Põldme et al., Chem. Sci. 10: 99-112, 2019
[3] L. A. Hammarback et al., Nat. Catal. 1: 830-840, 2018
[4] A. Bhattacherjee et al., Nat. Commun. 10: 5152, 2019

Pump-pump-probe spectroscopy sheds light on key reactions for artificial photosynthesis (ISMO, France)

Artificial photosynthesis aims at producing energy-rich chemical compounds, such as H_2 or methane, from H_2O and CO_2 using sunlight. Several steps have to be reproduced: sunlight must be captured, and electronic excitation must be converted into electronic energy stored in the catalyst using both charge separation and chemical



Schematic diagram of the photochemical process, along with a graph of the transient absorption spectrum of the pump-pump-probe pulse used to trigger it.

reactions. The main reactions that need to be mimicked are gathered into the oxygen evolution reaction (OER), the hydrogen evolution reaction (HER) and the CO_2 reduction reaction (CO_2RR).

These photocatalytic reactions, which produce solar fuels, require coupling of multiple photo-induced single-electron transfers (to accumulate charges on the catalytic centre) with multi-electron catalysis. Understanding of these light-driven charge accumulations in photosystems is thus of pivotal importance to guide optimisation of the photosystems. It has been identified as one of the bottlenecks to progress on this solar energy storage technology. A new experimental set-up at Institut des Sciences Moléculaires d'Orsay (ISMO) performs pump-pump-probe transient absorption spectroscopy to access the electronic excited states and their relaxation dynamics from the nanosecond to the millisecond range of timescale, in particular those of the charge accumulation and recombination in multicomponent molecular systems.

This set-up recently demonstrated light-induced two-electron accumulation on a model multicomponent system made of the $[Ru(bpy)_3]^{2+}$ molecule as the light absorber, ascorbate as the reversible electron donor and naphthalene diimide as the two-electron acceptor. Accumulative charge separation was observed in both single-pulse pump-probe and double-pulse pump-pump-probe experiments. The doubly reduced state was produced with up to 4.2 % yield and revealed a lifetime of about 100 microseconds. As far as it can be ascertained, this is the first demonstration of charge accumulation with only a single-pulse excitation in a multicomponent system. Time resolved resonance Raman spectroscopy is also being developed to complement the transient absorption experiments performed in the visible range of wavelengths. This type of investigation is key to mapping the path to artificial photosynthesis.

Performing such experiments in the presence of the substrate, CO_2 for instance, it is now possible to investigate the fundamental processes at work in the photocatalytic cycle of specific catalysts for laboratory-scale production of solar-derived fuels. It is noteworthy that one major challenge concerns the development of noble-metal-free, efficient, selective, and recyclable homogeneous or heterogeneous catalysts. Within the framework of collaborating investigation teams, synthesising photocatalysts in respect to this challenge, new families of hybrid organic-inorganic molecular photosystems are investigated for CO_2 reduction (CO_2RR) and HER reactions toward solar fuels.

Thomas Pino and Minh-Huong Ha-Thi (ISMO)

T. Tran et al., J.Phys.Chem. C 123: 28651, 2019

How to improve the efficiency and stability of organic dyes for luminescent solar concentrators (LENS, Italy)

Due to their high efficiency (15 – 20 %) and light-harvesting capability, silicon-based solar cells are widespread devices and presently contribute more than 2 % to global electricity generation. In the search for new technologies to boost the output of PV (photovoltaic) cells, the idea of increasing the light intensity by concentration is a potential solution to reduce the required photovoltaic material. Luminescent solar concentrators (LSCs) are a technology in which part of the spectrum of incident sunlight is down-converted by an efficient fluorophore dispersed in a transparent panel and sent to the surface of a PV cell. Due to the difference in refractive index between the panel material and air, most of the light emitted by the fluorophore is concentrated at the edges of the panel, where PV cells can be positioned. Since this concentrates the incident light by a factor of 5-10, without the need for expensive tracking, smaller silicon (or other) solar cells can be used. As the cost per square centimetre of the transparent plastic is lower than that of the solar cell, the cost per watt is lower than the cost of a planar silicon solar cell. Also, LSCs are of special interest for building integrated PV applications (for example in windows and roofs).

LSCs still have some drawbacks limiting their applications, particularly durability. Big efforts have been invested in developing new fluorophores and in particular, in designing suitable molecules with a wide absorption range (between 450 and 700 nm), high fluorescence quantum yield, large Stokes shift, good solubility in the host polymeric matrix and long optical and thermal stability. Various materials have been proposed and tested as fluorophores: quantum dots, inorganic and organometallic molecules, and small organic molecules such as perylene diimides, BODIPY oligomers, benzothiadiazoles and diketopyrrolopyrrole dyes.

Within this framework, the Ultrafast Spectroscopy group of the Photonic Materials area at LENS, the European Laboratory for Non-Linear Spectroscopy, started new collaborations with industrial laboratories and the Italian National Research Council (CNR), making use of standard spectrophotometric and laser techniques in characterising new organic molecules, aiming to identify the principal parameters making them usable in long term stable LSCs. Of course, materials that have facile and scalable synthetic routes, good processability, and high reproducibility are preferred.

Research efforts have been focussed on organic dyes with improved stability and efficiency in terms of fluorescence quantum yields and possessing a large Stokes shift. A combined approach of quantum mechanical calculations and photophysical characterisation of dye molecules has resulted in good strategies to achieve efficient fluorophores.

© Paolo Foggi

A piece of LSC material, showing increased brightness at the hand.

An analysis of the investigated organic fluorophores shows that a high radiative rate constant, that is, a high photoluminescence quantum yield and a short fluorescence lifetime, is one of the key points. In addition, isomerisation has been shown to worsen the photophysical characteristics of LSCs. Many fluorophores showing good characteristics in terms of efficiency and stability work by charge-transfer transitions. Rigidity of the molecular scaffold or of the surrounding polymer chains can help in increasing stability and making a more robust device. Pump-probe spectra with sub-picosecond pulses suggest that preventing large excited-state conformational relaxations, as happens in the polymeric matrix, results in increased fluorescence quantum yields.

Many of the investigated organic compounds belong to the donor-acceptor and donor-acceptor-donor type of molecules. They are synthetised to possess an intense emission spectrum between 450 – 600 nm, overlapping the light-absorption spectrum of Si-based photovoltaic cells. They down-convert the blue part of the solar spectrum where the efficiency of Si-cells is usually low. This causes the panel to be yellow-red coloured. The Stokes shifts are large (70 – 100 nm) to minimise the self-absorption effects. Recent results confirm the possibility of achieving efficiencies of more than 5 %.

Paolo Foggi (LENS), Mariangela DiDonato (LENS and CNR-ICCOM), Barbara Patrizi (LENS and CNR- INO)

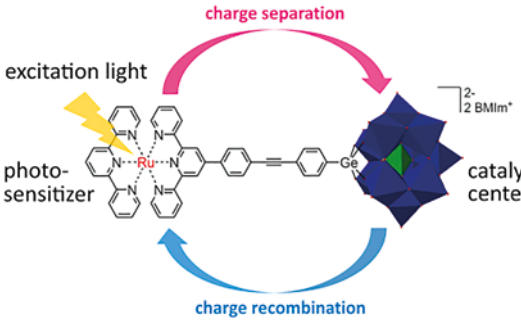
Using simulation, experiments and spectroscopy to improve the design of photocatalysts (IPHT, Germany)

Green plants and many algae obtain their energy from the conversion and storage of sunlight. In natural photosynthesis, light harvesting molecules such as chlorophylls absorb solar energy and convert it into one of nature's currencies of chemical energy, i.e. adenosine triphosphate (ATP). The plant uses this chemical energy and carbon dioxide (CO_2) to produce energy-rich molecules such as sugars, which can be stored over a long time.

Imitating what seems to work effortlessly in nature is still a great challenge for science. Based on the model of natural photosynthesis, the 2018 established Transregional Collaborative Research Centre (TRR 234) CataLight (Light-driven Molecular Catalysts in Hierarchically Structured Materials – Synthesis and Mechanistic Studies) researches novel soft-matter systems to convert solar energy into chemical energy carriers such as hydrogen – a renewable and clean fuel. To achieve this, CataLight embeds molecular photocatalysts, the mode of action of which can be tuned by molecular design, into soft-matter photocatalytic systems for the production of hydrogen and oxygen from water, but also for other industrially relevant photooxidation and photoreduction reactions [1].

The approach CataLight pursues is to integrate molecular photocatalysts into stimuli-responsive soft matter matrices and to understand the molecule-matrix interactions. The heterogeneous matrices offer vast opportunities to synergistically control charge transfer processes, photochemical reaction pathways and degradation resistance. However, the underlying mechanisms, which control component interactions, photochemical and -physical properties and overall catalytic activity are not fully understood yet. In order to make artificial photosynthesis a reality, these basic processes and mechanisms must first be elucidated.

Lasers are versatile and powerful tools to shine light on the complex light-activated multi-step reaction mechanisms that take place in a molecular catalyst for artificial photosynthesis. For example, in ultrafast time-resolved spectroscopy, pulsed lasers allow to picture processes that otherwise could not be observed because they take place on the pico- to femtosecond timescale and involve highly transient reactive intermediates. Combined with other laser-based techniques such as resonance-Raman spectroscopy, and computational studies, it is possible to deduce important structure-function-activity relationships that hence allow for the design of more efficient catalysts for artificial photosynthesis [2][3].



© Linda Zedler/Leibniz-IPHT, adapted from [2]

The kinetics of light-induced charge separation and recombination in a photocatalyst determine its efficiency for artificial photosynthesis.

Such a combined experimental, theoretical and spectroscopic approach was recently used to identify and spectroscopically characterise those highly reactive intermediates and their excited-state dynamics. Spectroelectrochemical methods (UV/Vis absorption and resonance-Raman spectroscopy), quantum-chemical simulations and time-resolved transient absorption (TA) spectroscopy elucidated the photoinduced electron transfer in the catalytically competent intermediate of a di-nuclear transition metal-based photocatalyst for hydrogenation of nicotinamide (NAD-analogue) and proton reduction. The in-situ photophysical studies on femto-to-nanosecond timescales revealed that electronic transitions shift electron density from the activated catalytic centre to a bridging ligand, which significantly reduces the catalytic activity upon visible-light irradiation. Based on these results, the photocatalysts can be synthetically re-designed to be more efficient – a spectroscopically guided approach to make artificial photosynthesis become reality.

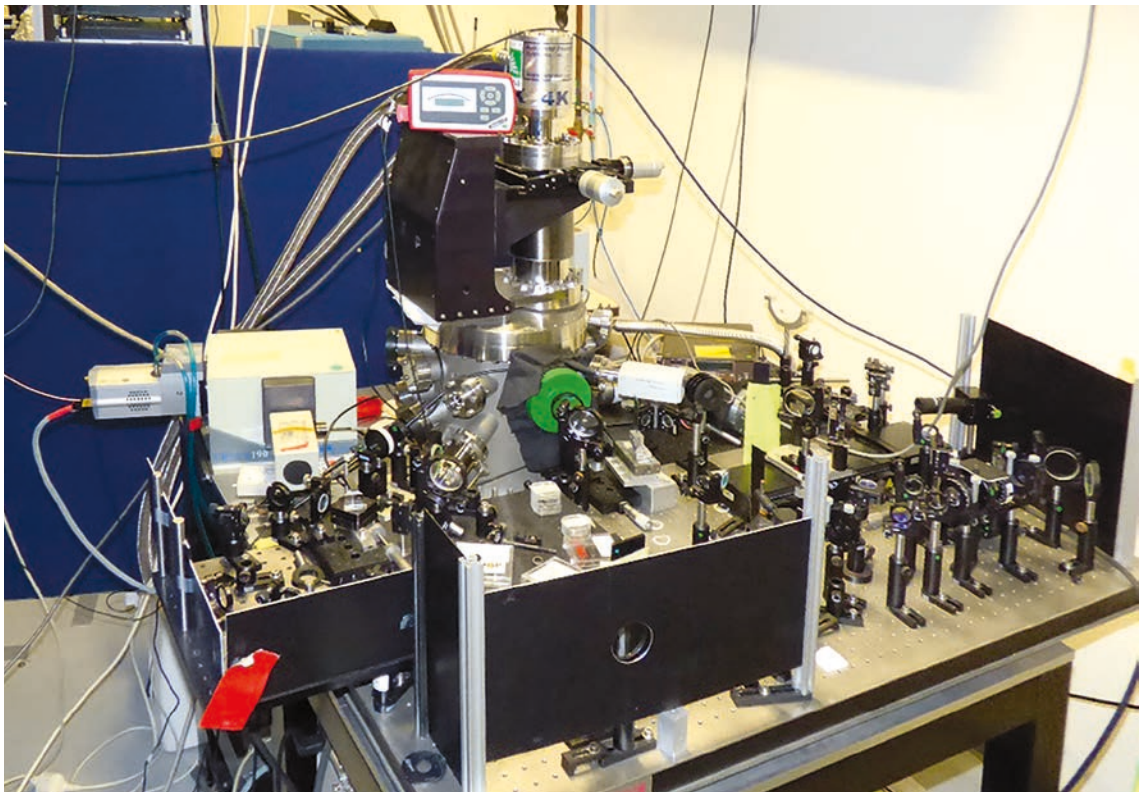
Benjamin Dietzek and Anja Schulz (IPHT)

[1] I. Krivtsov et al., Angew. Chem. Int. Ed. 59: 487, 2020
[2] L. Hammarström, Acc. Chem. Res. 48: 840, 2015
[3] F. A. Black et al., Chem. Sci. 9: 5578, 2018

Distant study of electronic dynamics in caesium-lead and organolead halide perovskites

The measurements for the transnational access project *Dynamics of electronic excitations in two lead halide perovskites with different cations: Cs^+ and methyl-ammonium group $CH_3NH_3^+$*, supported by Laserlab-Europe (project CNRS-CELIA002677), had initially been planned for May 2020 at CELIA (Bordeaux). Due to travel limitations induced by the COVID-19 pandemic, the visit had to be cancelled. However, the efforts of the CELIA staff members, who suggested that the experiments be performed remotely by the Russian PI and colleagues, transformed these limitations into a win-win situation. While the delivery of the samples from Russia to Bordeaux was a separate, non-trivial task, the experiment could be performed in October 2020 and all planned measurements were accomplished thanks to the scientific expertise of the CELIA group and previous experience of the Russian team at the host facility. Luminescence spectra and kinetics as a function of excitation density were investigated for a series of halide perovskites using a luminescence z-scan technique in the temperature range from LHeT to RT.

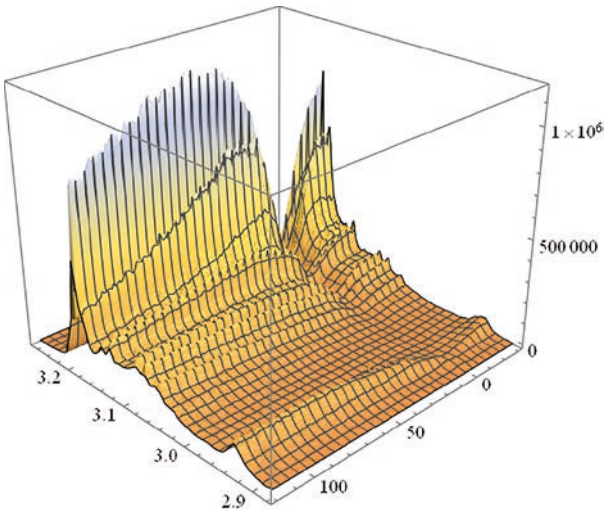
Their excellent optoelectronic properties and low production costs make metal-halide perovskites extremely promising materials in light harvesting and light emission for solar photovoltaics and LEDs, as can be seen from almost 10,000 papers dealing with halide perovskites indicated in the Scopus database for 2020 at time of writing. Superfast transformation of ionising radiation into light is another attractive feature. The origin of these properties lies in the specific electronic structure of these materials, with remarkably strong hybridisation of lead and halide states. The interband transitions at the onset of fundamental absorption appear to be also strongly coupled to excitonic transitions in lead-halide complexes. The small effective mass of the charge carriers results in their large mean free path, which is optimal for photovoltaic applications, and the high oscillator strength of the excitonic transition promises a fast sub-nanosecond fluorescence under photo- and gamma excitation. This rapid fluorescence allows the material to reach the ambitious goal of creating scintillator detectors with a resolution of tens of picoseconds. This would lead to the improvement of spatial resolution in time-of-flight medical positron-emission tomography as well as to super-high time resolution for future supercolliders of high luminosity in high-energy physics.



Luminescence beamline of the Aurore laser at CELIA.

Perovskites have also some other unique properties with substantial non-linearity of their luminescence yield as well as of spectral and kinetic parameters on excitation density. These characteristics are extremely important for simulating a scintillator response to ionising quanta/particles formed in different regions of their tracks with highly inhomogeneous excitation densities. They affect scintillation yield, energy proportionality, kinetics, and energy resolution, which is why they defined the scope of the present study. To change the density of excitations in the sample luminescent z-scan was used as the main technique, when the focus of the pulses of the 3rd harmonics (4.65 eV) of the femtosecond Ti-sapphire laser were shifted along the beam by the lens. With different pulse energies (from 1 to 500 nJ) and distances to the focal point the number of electronic excitations per cubic centimetre could be changed from 10^{16} to 10^{21}. An example z-scan is presented in the figure on the right. One can see that the luminescence spectra can be very different depending on the excitation density (lens position in this case), which also holds for kinetics, which were measured by the photomultiplier with microchannel plate. Zero of the z-axis corresponds to the focal point. Different energy transfer channels to the respective luminescence centres will be recovered by further analysis. One of the aims of the project was the comparison of inorganic materials like $CsPbCl_3$ with hybrid ones, in which caesium is replaced by methylammonium CH_3NH_3. Neither caesium nor CH_3NH_3 participate in the formation of luminescence centres, altering only lattice constants and details of the crystal structure with several phase transitions in the studied temperature range. Data for chlorides, bromides and mixed chloride-bromides obtained during this experiment are being prepared for publication.

Undoubtedly, it was a successful run and the highly professional CELIA team providing the laser beam and conducting the experiment guaranteed its success. Under the present circumstances, a remote experiment was the only possible approach, with both pros and cons. The benefits included the almost immediate online access to the results of the experiments for the project team, which was extended to the students of the Physics Faculty of Moscow State University, the regular discussions with French colleagues, the corrections to the measurements program during at least two daily on-line conferences. The obvious cons are the extensive load on the CELIA team and the absence of a Bordeaux ambience. The project authors are extremely grateful both to the CELIA team and for the support of Laserlab-Europe. This beamtime was a big step towards science without frontiers.



$CH_3NH_3PbCl_3$ luminescence spectra, excited by the 3rd harmonics of a femtosecond Ti-sapphire laser with 20 nJ/pulse at 13 K: The free and impurity-bound exciton luminescence were varied with the lens focal position.

Patrick Martin, Nikita Fedorov, Andrei Belsky (CELIA) and Irina Kamenskikh, Andrey Vasil'ev (M.V. Lomonosov Moscow State University)

IN MEMORIAM

Laserlab-Europe mourns the loss of its long-time colleagues David Neely, Theodore Papazoglou and Antonio Lucianetti

David Neely

Professor David Neely has passed away in August, after a short and sudden illness. He was an internationally renowned scientist in the field of high energy density science, and a highly valued collaborator in the Laserlab-Europe community. He had been working at the Central Laser Facility, UK, since 1993.

Theodore Papazoglou

Eminent scientist and researcher Theodore Papazoglou has died at the age of 57 after contracting the coronavirus. Before becoming head of unit at the Brussels-based European Research Council (ERC), he had worked at Laserlab-Europe partner Institute of Electronic Structure & Lasers, FORTH, Greece, for more than 11 years. He helped to set up the ERC and had served it for many years.

Antonio Lucianetti

Dr. Antonio Lucianetti, laser scientist at the Laserlab-Europe partner HiLASE, Czech Republic, has passed away at the age of 55, after a short illness. He had been a key member of the HiLASE team since the start of the project in 2011. He led Research Programme 2, whose Bivoj laser system set a world record for the first KW laser source in its class.

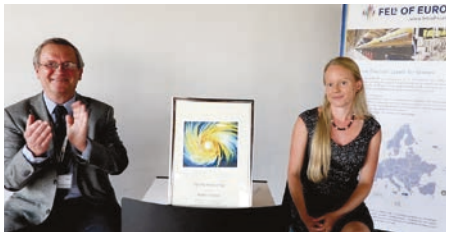
The Photon and Neutron Open Science Cloud (PaNOSC)

PaNOSC (the Photon and Neutron Open Science Cloud) aims to make Findable, Accessible, Interoperable and Reusable (FAIR) data a reality at six Photon and Neutron (PaN) research infrastructures. It promotes FAIR data management practices and is developing innovative data analysis services connected to the future European Open Science Cloud. PaNOSC collaborates closely with ExPaNDS (European Open Science Cloud Photon and Neutron Data Service). Both Horizon 2020 projects are working to make data from PaN facilities as open as possible and give access to data analysis services available remotely from a virtual portal. This effort is meant to benefit user communities and society at large by making science more open and reproducible, speeding up access to results and re-use.

PaNOSC and ExPaNDS have already developed FAIR-compliant data policy frameworks for PaN facilities and are developing a data search and analysis portal. The latter will give access to facility-specific dataset and data analysis services. An e-learning platform (https://pan-learning.org) for data-intensive training is also in production, aiming to provide a central point for developing and indexing training content and events.

ELI recently hosted an online "European PaN EOSC symposium" during which both projects gave updates on their status and plans. High-level representatives of ESFRI, EOSC, EOSC-Hub and the European Commission discussed PaN user perspectives on the future European Open Science Cloud. Presentations and videos are available on www.panosc.eu.

FELs of Europe organise first virtual "Science@FELs"



© DESY

R. Boll and European XFEL Scientific Director S. Molodtsov (left).

The Science@FELs conference is organised biennially by the "FELs of Europe" collaboration and has developed into a general conference in free-electron laser (FEL) science, highlighting scientific progress made in this field. In 2020, Laserlab-Europe co-organised the event for the third time. With a record of more than 700 registered participants for this first digital conference, held in September 2020, Science@FELs featured a lively exchange within nine invited sessions across the breadth of FEL science and applications.

Presentation topics included imaging, materials science, magnetic and correlated materials, femto-chemistry, catalysis, atomic and molecular physics, bioscience, and laser physics. The conference was scheduled for the European afternoon, which permitted Americans to attend early in their morning, while Asian colleagues participated during their late evening and night.

The conference was accompanied by three focus tutorials led by eminent FEL scientists, a virtual tour, and two "virtual poster sessions". While the poster session was a novel experience for nearly everybody, fruitful discussions took place and participants enjoyed the event.

During the virtual tour through FLASH and European XFEL facilities, participants had a behind-the-scenes experience with a live stream from the FLASH accelerator tunnel, experimental halls and laser hutches as well as special insights into all instruments at XFEL in dedicated videos and Q&A sessions.

This year's "FELs of Europe prize", recognising recent work of scientific excellence in FEL science and applications, was awarded during the conference to Dr. Rebecca Boll (European XFEL) "for her outstanding research on multiple ionisation of rare gases and photoinduced dynamics of ring-type molecules".

How to apply for access

Interested researchers are invited to contact the Laserlab-Europe website at www.laserlab-europe.eu/transnational-access, where they find relevant information about the participating facilities and local contact points as well as details about the submission procedure. Applicants are encouraged to contact any of the facilities directly to obtain additional information and assistance in preparing a proposal.

Proposal submission is done fully electronically, using the Laserlab-Europe Proposal Management System. Your proposal should contain a brief description of the scientific background and rationale of your project, of its objectives and of the added value of the expected results as well as the experimental set-up, methods and diagnostics that will be used.

Incoming proposals will be examined by the infrastructure you have indicated as host institution for formal compliance with the EU regulations, and then forwarded to the Access Selection Panel (ASP) of Laserlab-Europe. The ASP sends the proposal to external referees, who will judge the scientific content of the project and report their judgement to the ASP. The ASP will then take a final decision. In case the proposal is accepted, the host institution will instruct the applicant about further procedures.

Laserlab Forum Contact

Sylvie Jacquemot
Coordinator – Laserlab-Europe
Email: *jacquemot@laserlab-europe.eu*

Laserlab-Europe Office
Daniela Stozno
Max Born Institute
Max-Born-Str. 2A | 12489 Berlin | Germany
Phone: +49 30 6392 1508
Email: *stozno@laserlab-europe.eu*

Editorial Team:
Rebecca Davenport, Julia Michel, Daniela Stozno
Email: *office@laserlab-europe.eu*
Layout: unicom Werbeagentur GmbH

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