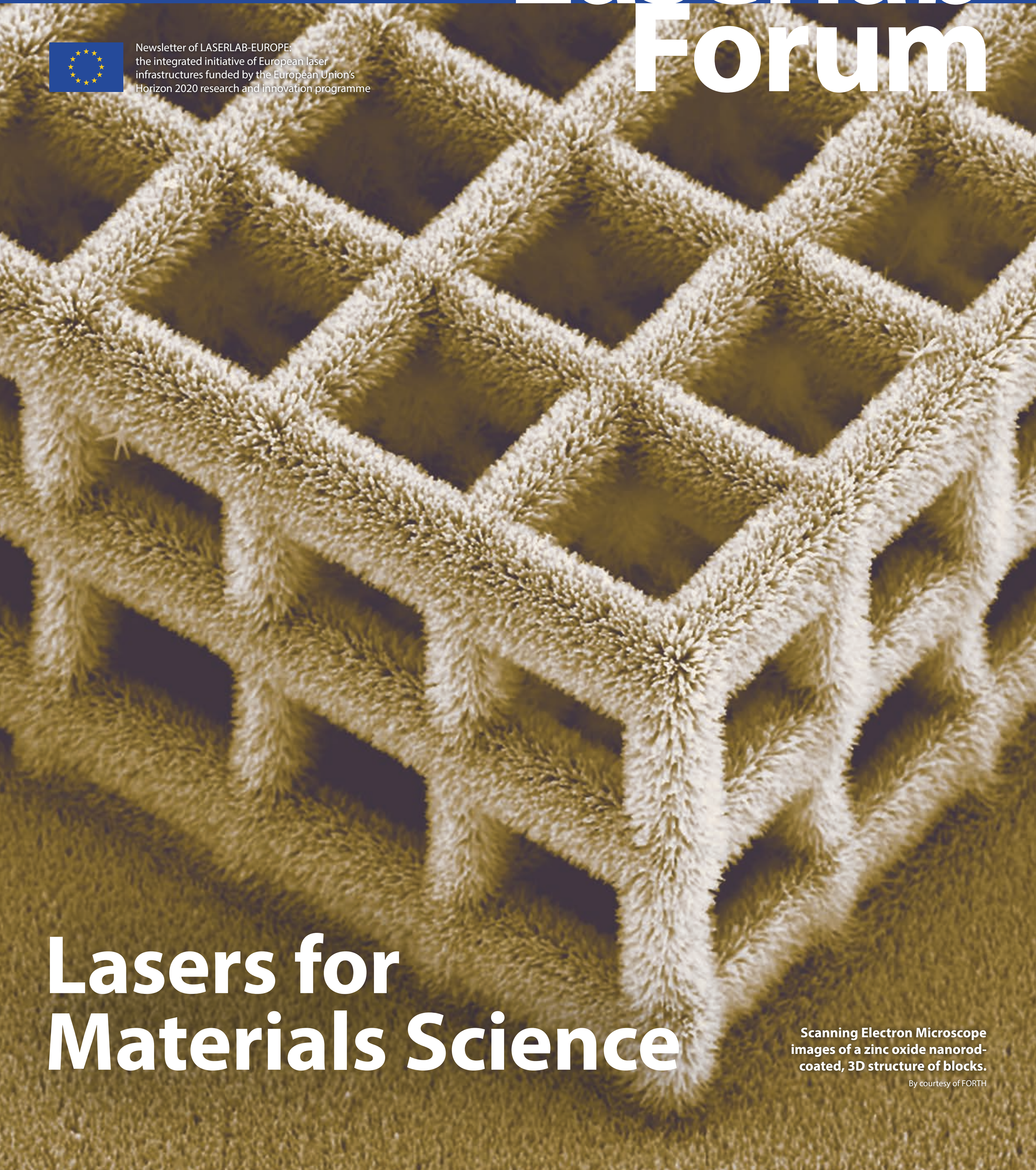
Issue 24 | December 2017

Laserlab Europe

Laserlab Forum

Newsletter of LASERLAB-EUROPE:
the integrated initiative of European laser infrastructures funded by the European Union's Horizon 2020 research and innovation programme



Lasers for Materials Science

Scanning Electron Microscope images of a zinc oxide nanorod-coated, 3D structure of blocks.
By courtesy of FORTH

Editorial

Tom Jeltes

It has been fifty years already since a laser was first used to cut through solid material. Back in 1967, Peter Houldcroft, working at The Welding Institute in Cambridge, got the luminous idea to try and use a 300 Watt CO_2 laser which was operational at the nearby Services Electronic Research Laboratory in Harlow to cut a 1 mm thick steel sheet. The experiment was successful, and laid the foundation for an entire industrial technology.

With the advent of ultrafast laser systems, a whole new register in laser 'cutting' technology has emerged. As can be read in this issue's focus section, laser ablation of materials is now used to make micro and even nanostructures – leading to designer materials with new properties. But the interest of lasers for materials science has a much broader scope than just the 'brute force' aspect of pulsed lasers – however sophisticated and precise those may be. Lasers are probably first and foremost excellent tools to study properties and behaviour of new and special materials, be it in the form of fluorescence or extreme ultraviolet microscopy, or as an essential ingredient in magnetic field imaging. All of these topics are highlighted in this issue's focus on Lasers for Materials Science.

Even the Access Highlight of this issue might be categorised under 'lasers for materials science', as it recounts a successful attempt to unravel the effect of gold nanoparticles on methane molecules, showing that gold might be the catalyst of choice if one wants to build long carbohydrate chains using methane as a feedstock molecule. That mechanism could turn out to be an efficient way of producing the future's organic materials.

Tom Jeltes

News

Laserlab-Europe members and associate partners meet

Laserlab-Europe held its annual General Assembly Meeting in October, this time in Bordeaux, France, hosted by CELIA (Centre Lasers Intenses et Applications). This year, directors of several associate partners of Laserlab attended the meeting: CALT, Croatia; LACUS, Switzerland; Laserlab DK, Denmark; Nanoscience Center, Finland; Orion, UK; and Wigner Research Centre for Physics, Hungary.

The event provided an excellent opportunity to learn about facility updates and development plans and highlighted a broad range of facilities of different scale and scientific focus. Together with subcontractors and associate partners, Laserlab-Europe covers activities in 23 European countries.

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A major topic of the meeting was the discussion about the creation of a formal legal entity in order to promote the sustainability of Laserlab-Europe and to allow for activities beyond the scale of an EU-funded project.

Time limit for ultrafast perovskite solar cells

In a transnational access project, researchers from Laserlab-Europe partner CUSBO in Milan, with colleagues from Cambridge University, have quantified the extremely high speeds at which future solar cells would have to operate in order to stretch what are presently seen as natural limits on their energy efficiency. The study, which was published in Nature Communications (8: 376, 2017), investigated photovoltaic devices based on perovskites, and suggests that these could achieve unprecedented efficiency if the generated electrical charge is extracted within femtoseconds.

When sunlight hits the cell, photons are converted into electrons. These can be drawn out through an electrode to harvest electrical charge. For a brief moment after they are created, the electrons are moving very quickly. However, they then start to collide, and lose energy. Electrons which retain their speed, prior to collision, are known as 'hot' and their added kinetic energy means that they have the potential to produce more charge.

The study found that electron collision events started to happen between ten and a hundred femtoseconds after light was initially absorbed by the cell. Thus, to maximise energy efficiency, the electrons would need to reach the electrode in as little as ten femtoseconds. According to the authors, it might be possible to achieve such extraction speeds in perovskite solar cells.

What is Laserlab-Europe?

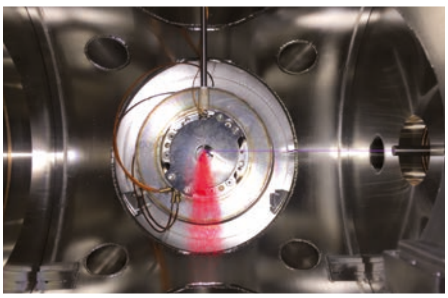
Laserlab-Europe, the Integrated Initiative of European Laser Research Infrastructures, understands itself as the central place in Europe where new developments in laser research take place in a flexible and co-ordinated fashion beyond the potential of a national scale. The Consortium currently brings together 33 leading organisations in laser-based inter-disciplinary research from 16 countries. 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.

Ultrashort photoemission time of electrons clocked

Researchers from Laserlab-Europe partner Lund Laser Centre (LLC), together with Swedish colleagues, have been able to measure the incredibly brief time it takes for electrons to be emitted from neon atoms, using attosecond laser pulses. With their ultraprecise experiment, they were able to solve an apparent discrepancy between theory and experiment found in 2010. Their results were published in Science (358: 893, 2017).

When an atom absorbs a photon, the energy can be used to emit an electron. This photoemission process does not happen instantaneously, as was shown by a team from the Max Planck Institute of Quantum Optics (MPQ) – also part of Laserlab-Europe – in a landmark experiment in 2010. More specifically, they found a difference between the emission times of electrons from different orbitals. The difference they measured, however, was more than twice as long as accounted for by theory.

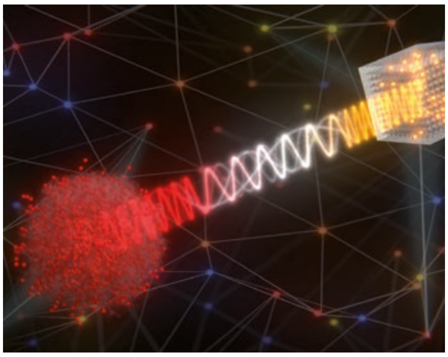
The Lund team, led by Anne L'Huillier, together with colleagues from Stockholm and Gothenburg, have now been able to solve this puzzle. Using a train of attosecond laser pulses of extreme ultraviolet light combined with an infrared laser, as well as sensitive electron detection which could distinguish between electrons with an energy difference of only 100 meV, they were able to show that the discrepancy arose from a so-called 'shake-up' of electrons. Here, one electron is emitted, while a second electron is promoted to a higher orbital – which accounted for the longer time differences measured at MPQ.



Inside the vacuum chamber

Quantum internet goes hybrid

A team of researchers from Laserlab-Europe partner ICFO (Barcelona) have reported the first demonstration of an elementary link of a hybrid quantum information network, using a cold atomic cloud and a doped crystal as quantum nodes and single photons as information carriers. Their work was published in Nature (551: 485, 2017).



Schematic illustration of a hybrid information network.

In their study, the ICFO researchers used two very distinct quantum nodes: the emitting node was a laser-cooled cloud of rubidium atoms and the receiving node a crystal doped with praseodymium ions. From the cold gas, they generated a quantum bit (qubit) encoded in a single photon with a very-narrow bandwidth and a wavelength of 780 nanometre. They then converted the photon to the telecommunication's wavelength of 1552 nanometre to demonstrate that this network could be completely compatible with the current telecom C-band range.

Subsequently, they sent it through an optical fibre from one lab to the other. Once in the second lab, the photon's wavelength was converted to 606 nanometre in order to interact correctly and transfer the quantum state to the receiving doped crystal node. Upon interaction with the crystal, the photonic qubit was stored in the crystal for approximately 2.5 microseconds and retrieved with very high fidelity.

High resolution without particle accelerator

Physicists from the Helmholtz Institute Jena (HIJ), partner of Laserlab-Europe, have for the first time been able to achieve optical coherence tomography with extreme ultraviolet (XUV) light at laboratory scale. The method developed in Jena, which relies on ultrashort laser pulses focussed into noble gases, opens up possibilities to relatively cheaply create high-resolution tomographic images. The results have been published in the journal Optica (4: 903, 2017).

In optical coherence tomography – an imaging process used by, e.g., optometrists – a shorter wavelength provides a higher resolution. Physicists at Friedrich Schiller University and the Helmholtz Institute Jena have now used extreme ultraviolet radiation (XUV) for coherence tomography, generated in their own laboratory. Usually, large-scale particle accelerators are necessary to create XUV radiation, but the Jena researchers found an alternative way by focussing an ultrashort, very intense infrared laser in a noble gas, such as argon or neon. The radiation has a wavelength of between 20 and 40 nanometres – from which it is just a small step to the X-ray range.

HIJ researcher Silvio Fuchs in a laboratory of the Institute of Optics and Quantum Electronics of the Friedrich Schiller University.

ERC Grants

Each year, a significant number of Laserlab-Europe researchers are awarded prestigious personal grants from the European Research Council. Here, we highlight four recently granted Starting and Consolidator projects – worth up to 1.5 and 2 million euros, respectively – for a period of five years.

Starting Grant Andreas Reiserer (MPQ): A scalable quantum network

A future quantum network will consist of quantum processors that are connected by quantum channels, just like conventional computers are wired up to form the Internet. While pioneering experiments have demonstrated the entanglement of two quantum nodes separated by up to 1.3 km, accessing the full potential of quantum networks requires scaling of these prototypes to more nodes and larger distances.

In his Starting Grant project QuantumNet, Andreas Reiserer proposes to use the exceptional properties of individual Erbium ions embedded in Yttrium crystals to increase the size of quantum networks, using the concept of quantum repeaters. To attain this goal, he intends to create a quantum spin-photon interface at a telecommunication wavelength, followed by multiplexing of many quantum bits in one device via frequency-selective addressing. Next, he will use remote entanglement swapping and purification to increase the range of quantum-secure communication beyond its current fundamental limit.

Starting Grant Simon Wall (ICFO): Fluctuations in high-temperature superconductors

One of the major open questions in condensed matter physics is the origin of high-temperature superconductivity. Electron interactions are considered responsible for this phenomenon, but despite over thirty years of research, experimental proof has been difficult to obtain.

With his Starting Grant project SeeSuper, Simon Wall aims to break this deadlock by applying new techniques to study the superconducting state. His strategy is to probe high-temperature superconductors through their nanoscale and femtosecond fluctuations. He will focus on three key parameters in superconductors: phonons, spins and nanoscale phase separation, with the aim of revealing the coupling mechanism.

Wall hopes to prove the hypothesis that lattice anharmonicity is the key missing ingredient to explain the origins of high temperature superconductivity. His experimental approach combines transient optical spectroscopy and time-resolved diffuse X-ray scattering to measure the lattice response to large amplitude coherent vibrations, time-resolved non-linear optical spectroscopy to directly probe spin dynamics, and resonant soft X-ray holography to image dynamics on the nanoscale.

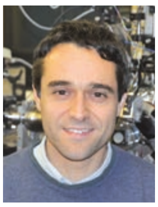
Consolidator Grant Tomáš Čižmár (IPHT): Holographic micro-endoscopy

To date, no high-resolution in-vivo imaging technique has been introduced which would be applicable deep inside living organisms without causing significant harm to the surrounding tissues. In this context, an important challenge is to increase the penetration depth of superresolution imaging inside living organisms.

In his Consolidator Grant project, Tomáš Čižmár therefore aims to develop new, ultra-thin endoscopic devices for acquiring high-quality images from unprecedented depths inside tissues of living organisms. For this purpose, assisted by a team of transdisciplinary experts, he intends to push the boundaries of the holographic control of light propagation in multimode fibres. The idea is that this approach will enable his team to deploy several light-based imaging methods, including superresolution microscopy, inside freely moving animal models and ultimately humans.

In the first instance, the micro-endoscopy technique will be applied to neuroscience, providing a minimally invasive window into fundamental processes in neuronal diseases such as Alzheimer's.

Consolidator Grant Nicola Poli (LENS): Gravitational effects on quantum systems



After 100 years of general relativity and quantum mechanics, both theories have been tested at an unprecedented level. Unfortunately, though, we lack full comprehension at the fundamental level: a quantum description of space-time is still under discussion. High-precision experiments capable of measuring tiny gravitational effects on quantum systems, such as new generation atomic clocks and quantum inertial sensors, might be used to observe effects originating from Planck-scale physics.

The main aim of Nicola Poli's Consolidator Grant project is therefore to explore the limits of contemporary physics with a new generation of atomic quantum sensors, namely optical atomic clocks and atomic gravimeters. He will perform ultimate precision tests of gravity with fountains of ultra-cold cadmium and strontium atoms. Specifically, he aims to perform tests of the weak equivalence principle with quantum probes, exploring also possible spin-gravity couplings. Furthermore, he will address quantum interference of high-precision clocks in a gravitational potential and will try to demonstrate gravity-induced decoherence mechanisms, opening the way towards an explanation of the quantum-to-classical transition in macroscopically entangled quantum systems.

EUCALL completes Mid Term Review and 2nd Annual Meeting

In June 2017, the European Cluster of Advanced Laser Light Sources (EUCALL) completed its Mid Term Review in Brussels, during which the EUCALL coordinator team and work package leaders presented the project's progress and achievements to the EU project officer and an external reviewer.

The review recognised that EUCALL (a Horizon 2020 project, grant agreement No 654220) has fully achieved its objectives and milestones for the first funding period and is creating a wealth of advanced results, instruments, and tools. The review specifically emphasised and supported EUCALL's compiled spreadsheet of "Instrumentation at Advanced Laser Light Sources", which contains characteristics of UV and X-ray instruments at synchrotron, free electron laser (FEL) and optical laser facilities. The spreadsheet was described as an "invaluable document", especially when the collected data will be transferred to the public www.wayforlight.eu database for users of facilities in the EUCALL community to identify the most suitable instruments/beamlines for a defined experiment. This will be of particular benefit to Laserlab-Europe, as selected instruments will appear on the database, which previously featured just synchrotron and FEL facilities.

The development of EUCALL's SIMEX tool was also highlighted as a significant result. SIMEX is a new open-source simulation platform, available through the EUCALL webpage, for users and facility operators to simulate experiments "from source to detector" at advanced light sources. It includes software separately developed and put together in one package for the simulation of experiments at the different light sources. SIMEX currently supports simulations of coherent diffractive imaging, as well as of X-ray imaging and scattering experiments on optical laser-excited or compressed matter.

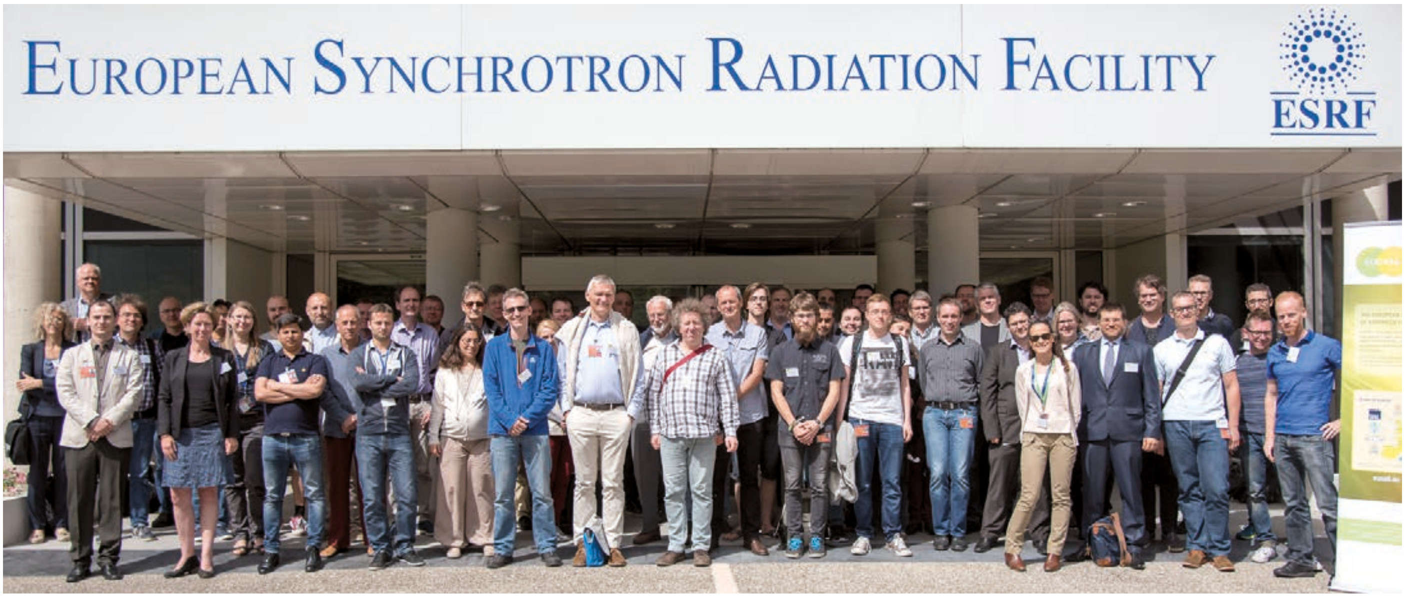
The reviewer also identified EUCALL's networking activities around newer concepts and approaches to be of great value for structuring an emerging community. For example, EUCALL organised a workshop on "Biology at Advanced Laser Light Sources" (30 Nov – 1 Dec 2017 at European XFEL). The focus on biology applications was selected since these are currently of very high relevance to all of the participating facilities. The workshop promoted understanding of the synergies between the different subgroups, as well as how to identify and develop them and later to extend these synergies into other communities.

A second workshop on "Societally Relevant Science at Advanced Laser Light Sources" – to be held at DESY on 26-27 April 2018 – will address urgent scientific and societal challenges and how EUCALL's facilities could contribute to solving these. A further topic deals with building a network for target delivery at high-repetition-rate laser facilities, to be discussed at a workshop at ELI-Beamlines during 28-30 May 2018.

In June 2017, EUCALL held its 2nd Annual Meeting at ESRF in Grenoble. Sixty-six project participants gathered to present and discuss the project progress and status. A plenary session addressed photon beam characteristics and applications of FEL, synchrotron, and various optical laser-driven X-ray beams and one session was dedicated to discuss possible collaboration after the EUCALL project ends in September 2018. In the open discussion that followed, the participants unanimously agreed to continue EUCALL's cooperation in the future.

Graham Appleby

www.eucall.eu



EUCALL's project participants gathered at the Annual Meeting 2017 at ESRF.

Lasers for Materials Science

Lasers can be used to study or manipulate materials in many different ways. This focus shows how researchers from several of Laserlab-Europe's partners are involved in laser-based materials science. The highlighted projects include diagnostic application of lasers, such as extreme ultraviolet light for ultraprecise microscopy, spectroscopy of photoactive materials, fluorescence imaging of a range of materials, and laser-assisted imaging of a material's magnetic field. Another contribution shows how ultrashort, high-power laser pulses can be used to study phase transitions in solids. More directed at the manipulation of materials are accounts on photolithographic fabrication of nanorods and precision laser ablation, as well as on the mechanisms behind the ablation process. These examples illustrate the variety of techniques for materials science within Laserlab-Europe, which are available for users from outside the consortium via the transnational access programme.

Laser-based fluorescence microscopy for materials science (CLF, UK)

Researchers in the Octopus lab at the Central Laser Facility.

The *Octopus* microscopy cluster at the UK's Central Laser Facility (CLF) offers most laser-based fluorescence microscopy techniques to the user community. Lately there has been increased interest from the physical sciences community in these traditionally biological imaging methods, promoted by the interdisciplinary environment of the facility in the Research Complex at Harwell (RCaH).

Recently, a research group led by Prof. Fiona Meldrum from Leeds University investigated the incorporation of organic additives into crystalline materials [1]. These additives are often used to control the crystallisation process. This work used a combination of scanning electron microscopy and X-ray diffraction to show the location of the occlusions and their effect on crystal morphology, while fluorescence spectroscopy and lifetime imaging on *Octopus* was used to probe the local environment of the occlusions within different zones of the crystal. The strategy was then extended to incorporate simultaneously mixtures of dyes, whose fluorescence cascade creates calcite nanoparticles that fluoresce white. This offers a simple strategy for generating biocompatible and stable fluorescent nanoparticles whose output can be tuned as required.

The location of the UK Catalysis Hub in RCaH has prompted a number of interactions with the CLF, including use of *Octopus*. In a project led by Prof. Andrew Beale (University College London/RCaH) [2] again a combination of X-ray and optical microscopy was used to investigate a physical sciences system. This work showed how molybdenum speciation and hydrocarbon accumulation affect the performance of catalysts used in the conversion of methane into high-value chemicals.

The final example is in the use of super-resolution imaging techniques for the characterisation of advanced materials. A study headed by Ian Manners (Bristol University) [3] used a combination of X-ray spectroscopy, electron microscopy, and super-resolution fluorescence microscopy to study platelet micelles. These structures are built from block copolymers and the solid and hollow 2D micelles provide a tunable platform for further functionalisation and have potential for a variety of applications.

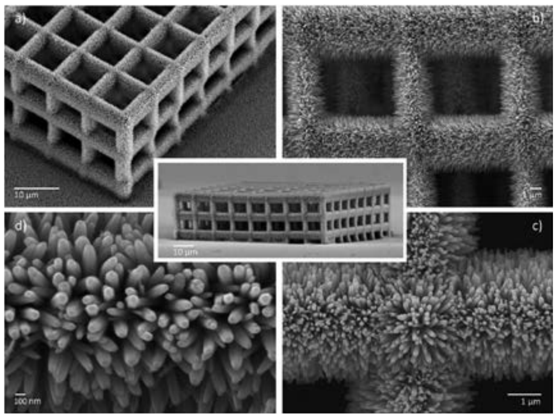
We expect continued growth in the use of these techniques for physical sciences research. The interdisciplinary nature of RCaH and its colocation with facilities offering complementary techniques makes it well placed to make the most of these developments. **Dave Clarke**

[1] D.C. Green et al., Nature Communications 7: 13524, 2016
[2] I. Lezcano-Gonzalez et al., Angew. Chem. Int. Ed. 55: 5215, 2016
[3] H. Qiu et al., Science 352: 697, 2016

Laser-made 3D zinc oxide nanostructures (IESL-FORTH, Greece)

Fabrication of 3D zinc oxide (ZnO) nanostructures is important for the development of novel ZnO-based devices, such as photocatalysts. Researchers from IESL-FORTH (Crete, Greece) have recently demonstrated that complex patterns of three-dimensional ZnO nanorods can be made combining nanolaser techniques with low-temperature hydrothermal growth.

Zinc oxide (ZnO) is a widely-studied metal oxide semiconductor, due to its potential use in a variety of applications, such as gas sensors, photocatalysts, nanolasers, photo-electrochemical cells for hydrogen generation from water splitting, and photoluminescent devices. Its useful properties, but also the various geometries that can be grown, such as nanorods and nanobelts, make it one of the most studied materials in nanoscience. For the fabrication of pure and doped ZnO nanostructures, several chemical and physical synthesis methods have been adopted to control the distribution of ZnO nanostructures on mainly flat surfaces.



Scanning Electron Microscope images of a zinc oxide nanorod-coated 3D structure of blocks (centre), fabricated with Multi-Photon Lithography and zinc-seeded aqueous chemical growth. Reproduced from Giakoumaki et al. 2017.

At IESL, researchers have recently developed an innovative method for the fabrication of fully 3D ZnO nanorod-coated structures, which involves seeded hydrothermal growth of ZnO nanorods on a 3D scaffold of an organic-inorganic hybrid material, fabricated by Multi-Photon Lithography (MPL). The growth of ZnO nanorods is based on a two-step procedure that requires the deposition of a metallic zinc seed layer onto the polymeric scaffold, employing the pulsed laser deposition (PLD) technique, followed by an aqueous chemical growth of ZnO nanocrystalline rods out of an aqueous solution of zinc nitrate hexahydrate ($Zn(NO_3)_2$) in the presence of ammonia.

This is a straightforward and flexible scheme carried out at relatively low temperature (<100°C), and is consistent with different types of substrates. Additionally, the laser-based techniques employed for the fabrication of 3D the scaffold (MPL) and the seed Zn layer (PLD) allow the deposition of ZnO nanorods in the form of micro-architected patterns on substrates with flat or complex geometry. Additional variability in the nanorod structure architecture can be introduced by varying the growth conditions, while the incorporation of appropriate chemicals would enable the growth of doped or functionalized ZnO nanorods.

Maria Farsari and Argyro Klini

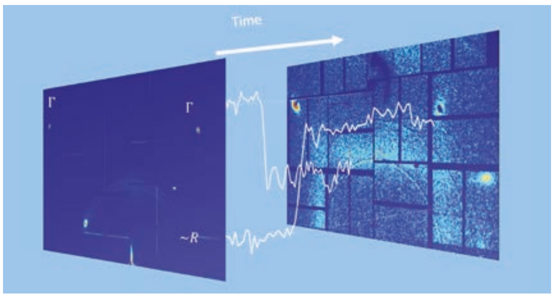
A.N. Giakoumaki et al., Scientific Reports 7: 2100, 2017

A new look at ultrafast phase transitions (ICFO, Spain)

Researchers from the Ultrafast Dynamics of Quantum Solids group of Dr. Simon Wall at ICFO, Barcelona, Spain, in collaboration with groups at the Pulse Institute at SLAC and Duke University in the USA, have used free electron lasers in the USA and Japan to watch the femtosecond dynamics of a phase transition in a new way.

Phase transitions in solids can be studied using Bragg reflection of X-rays, which reflect from samples at specific angles due to constructive interference between planes of ordered atoms. The higher the crystallinity, the sharper and more intense the reflectivity, or Bragg peaks, appears. If a phase transition occurs, some symmetry planes may be lost, resulting in a change in the number and position of the Bragg peaks.

Researchers now routinely exploit this fact to measure how phase transitions occur in real time. Femtosecond pulses of light are used to trigger a phase transition and the intensity of the Bragg peaks are monitored with pulsed X-ray sources. However, whilst this technique measures the timescale of phase transitions, it does not reveal how the transition occurs.



Evolution of the diffuse scattering during a photoinduced phase transition in vanadium oxide (VO_2).

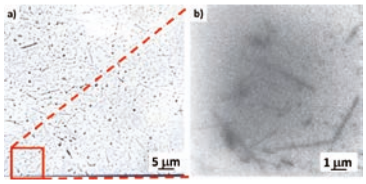
To overcome this limitation the researchers used time-resolved thermal diffuse scattering (TDS) of the structural transition in vanadium oxide (VO_2). This technique measures the inelastically scattered X-rays from a sample, which is typically orders of magnitude weaker than the elastically scattered X-rays, but is sensitive to the displacements of the atoms from their average positions. With the development of ultrafast and ultrabright X-ray free electron laser sources this signal can now be measured in the time domain.

The TDS signal showed that, while the lattice initially makes a cooperative motion after photoexcitation, within 60 femtoseconds atomic motions become random, which ultimately drives the phase transition. The ultrafast nature of this transition from coherent to incoherent motion provides a new interpretation of the phase transition in terms of rapid transfer of energy between vibrational modes of the system, and points to strong anharmonicity as a mechanism for providing the rapid switching time.

Simon Wall

Extreme ultraviolet (EUV) microscope for imaging of nanostructures (MUT, Poland)

A compact, desktop-sized microscope, based on a laser-plasma light source and a diffractive Fresnel zone plate objective, was developed by Polish Laserlab-Europe partner IOE-MUT. The microscope operates in the extreme ultraviolet (EUV) region at the wavelength of 13.8 nanometre, and has been used for imaging of silver nanowires.



Images of the silver nanowires obtained for comparison between a visible light micrograph, obtained with a NA = 0.7, 40x objective (a) and the EUV microscope (b). To acquire the EUV image, 200 EUV pulses were accumulated, at a 10 Hz source repetition rate. In the images nanowires with variable lengths and thicknesses of the order of a fraction of a micron can be observed. Reproduced from Torrisi et al. 2017.

Progress in materials science, especially in nanoscience and nanotechnology, requires improvement of the spatial resolution of microscopes in order to obtain images on the nanometre scale. Light of shorter wavelength can be focused into smaller volumes, improving the diffraction limited resolution and allowing to see finer details. Imaging with radiation in the extreme ultraviolet (EUV) spectral range (10 to 121 nm) has been demonstrated using various light sources, including synchrotrons and free electron lasers (reaching a spatial resolution of ~10 nm), as well as light sources based on plasmas and high-order harmonics generation (yielding sub-100 nm resolution).

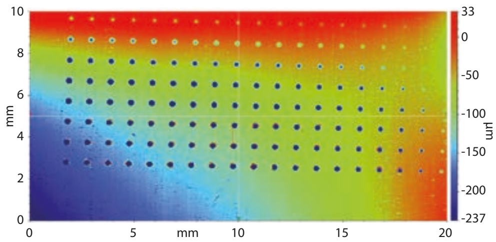
Researchers from the Institute of Optoelectronics-Military University of Technology (IOE-MUT) have now developed a compact, desktop-sized microscope using a laser-plasma EUV source and Fresnel optics. The source is based on an argon gas puff target irradiated with nanosecond pulses from a commercial Nd:YAG laser. The microscope is operating at the wavelength of 13.8 nm, which is selected from the argon plasma emission using an ellipsoidal off-axis collector with Mo/Si multilayer coating. EUV-illuminated samples are imaged in transmission mode onto a CCD camera using a Fresnel micro-zone plate. The spatial resolution obtained from measurements performed using a well-established 'knife-edge' test was about 95 nm.

The EUV microscope was used for imaging of silver nanowires deposited on top of a 30 nm thick silicon nitride membrane. The nanowires, with a diameter in the range of 10-100 nm and a length of a few micrometers, were synthesized using $CuCl_2$-mediated polyol process. The EUV image exhibits superior spatial resolution and much higher optical magnification (410x) compared with a visible light micrograph.

Henryk Fiedorowicz and Przemyslaw Wachulak

A.Torrisi et al., Journal of Microscopy 265: 251, 2017

Pulse shaping for improved femtosecond ablation (IST, Portugal and CLPU, Spain)



Two-dimensional map of the depth profile of holes in copper. The colour scale shows depths in micrometres. Number of shots ranging from 10 to 5000 and energy from 25 μJ to 2.7 mJ.

Over the past two decades, femtosecond laser pulses have been used for probing delicate living structures without damaging them, or for high precision microprocessing of high quality materials on the micro- and nanoscale. Researchers at IST in Lisbon and CLPU in Salamanca have now teamed up to study the ablation by custom-shaped pulses on metallic and organic targets.

Laser ablation is a material processing technique that has undergone a number of technological advances in recent years. The use of extremely short (femtosecond level) laser pulses is at the forefront of many fundamental applications, as shorter driving laser pulses lead to a higher quality material processing.

The laser teams in Lisbon and Salamanca, led by Dr. Gonçalo Figueira and Dr. Mauricio Rico, respectively, have joined their efforts and experimental capabilities in a new collaboration. Their goal is to investigate the mechanism of ultrafast laser ablation for biological applications by employing simultaneous spatial and temporal shaping of the laser beam. This technique holds the potential to significantly improve the efficiency of the process, contributing to a better understanding of the material processing methods at such extreme timescales.

The Portuguese team has developed an automated experimental setup for spatial shaping of the laser pulse. Preliminary studies were concentrated on metals because of their homogeneous structure and well known properties. Additionally, copper and teeth samples were tested.

The main results from this ongoing work are the determination of the thermal threshold fluence – the laser energy density at which the target is vaporised – for two significantly different types of materials (organic and metallic), in the femtosecond pulsed regime. The conditions for micro-crack formation and strong damage induced to the surrounding material were also found. Besides these achievements, the depth profiles of ablated holes and lines and their dependence on fluence were studied.

The collaboration with CLPU arose from the expertise developed by this team in several aspects of laser ablation, namely the sample preparation, interaction and analysis. IST researcher Victor Hariton acquired practical know-how at their facilities in the scope of a Laserlab-Europe staff exchange grant.

The next goals include developing the temporal pulse shaping setup in order to evaluate the ablation rate and compare the results with analytic calculations.

Victor Hariton, Mauricio Rico and Gonçalo Figueira

Mechanisms of laser-induced ablation in dielectrics (SLIC, France)

Femtosecond lasers can be efficiently used to drill, ablate, cut, or to permanently modify – in three dimensions – the optical properties of transparent materials, giving rise to numerous applications. Improving laser processing requires a detailed knowledge of the physical mechanisms involved during the interaction. To get more insight in this complex situation, experiments have recently been carried out on silica and sapphire at SLIC, Saclay.

Understanding the physics underlying laser damaging, breakdown, or processing of wide band gap dielectrics, is a challenging task due to the competition between many different elementary physical mechanisms, all occurring at sub-picosecond or femtosecond time scale: electron-phonon interaction, elastic and inelastic electron-electron scattering – including impact ionisation, formation of transient or permanent defect states, exciton self-trapping, exciton-exciton interaction, etc.

Recently, in a transnational access project, experiments have been carried out in Saclay (SLIC facility) on silica (SiO_2) and sapphire (Al_2O_3) samples, in collaboration with two teams from Aarhus University (Prof. Peter Balling) and Madrid (Prof. Javier Solis). Modification of the optical properties of the surface and the bulk was measured simultaneously using time-resolved reflectivity and Fourier-transform interferometry, at pump intensity up to three times the ablation threshold, over a temporal span of 300 picoseconds. The main outcomes of these investigations are the following:

For both materials, silica and sapphire, and for fluences well above the ablation threshold, the maximum measured plasma reflectivity shows a saturation behaviour. Our numerical simulations show that in this high fluence regime, for the pulse duration used (120 fs FWHM), collisional excitation dominates over strong-field excitation. This causes a cooling of the conduction band electrons down to the Fermi velocity limit, where the carrier-carrier scattering rate becomes proportional to the density of free electrons. As a consequence, the reflectance of the excited surface becomes nearly constant according to the Drude-Lorentz model.

Finally, in the temporal window of 10–30 picoseconds, both materials simultaneously display low reflectivity (close to zero) and high absorption (close to one). This suggests that, before the excited region substantially expands or exchanges heat with the surroundings, the material transiently behaves optically like a high-temperature blackbody.

Stéphane Guizard

M. Garcia-Lechuga et al., Phys. Rev. B 95: 214114, 2017

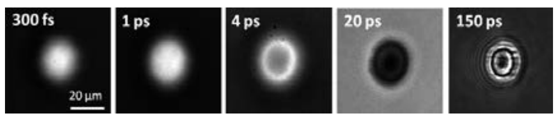


Image of reflected probe beam at the surface of α-Al_2O_3, for different time delays.

Magnetic field imaging using nitrogen-vacancy centres in diamond (ULLC, Latvia)

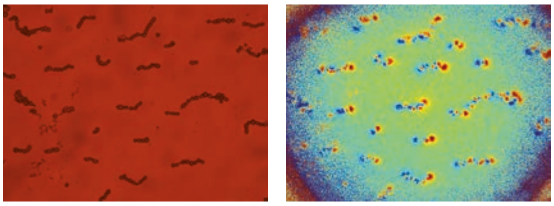
A recent study by a team from the Laser Centre of University of Latvia (ULLC) shows that magnetometry based on nitrogen-vacancy centres can be used to create microscope-like images of the magnetic field of small magnetic particles, with unrivalled spatial resolution. With the advent of inexpensive high-power green diode lasers, this magnetometry method has now become an inexpensive imaging tool.

An image of the magnetic field distribution can offer insights beyond what could be obtained purely through optical microscopy when dealing with paramagnetic materials. Magnetotactic bacteria, magnetorheological materials or magnetic labelling materials all have magnetic features of micrometre scales. However, magnetic measurement devices based on alkali metal vapour cells, SQUIDs or Hall probes either lack the spatial resolution to discern the tiny features or their operating conditions are such that close proximity could adversely affect the sample.

As part of Laserlab-Europe's Joint Research Activity PHOTMAT, researchers from ULLC studied nitrogen-vacancy centres in diamond as a tool for magnetic imaging. Nitrogen-vacancy centres are point defects in the lattice of diamonds, where two nearest-neighbour carbon atoms are replaced by a nitrogen atom and a hole (vacancy). This produces an atom-like colour defect in a triplet spin state, which can be polarized and read out optically. The defects are commonly formed by ion implantation and annealing, allowing one to engineer a diamond chip with nitrogen-vacancy centres 100 nanometres beneath the surface, ensuring close proximity between the sensor and the sample.

Using a combination of optical pumping with a green laser and an applied microwave field, the local magnetic field inside the diamond chip, as produced by small magnetic sample particles placed on its surface, can be deduced from the spatial variation of the fluorescence emitted by the nitrogen-vacancy centre in a method called optically detected magnetic resonance (ODMR).

The purpose was to obtain images of magnetic field distributions for particles whose magnetic moment have been measured by other means, and to compare these measurements to the values obtained from the magnetic field images. The ULLC researchers were able to detect particles as small as 19 nanometres, though the optical resolution is still diffraction limited (~500 nm). They conclude that while the imaging method is definitely less sensitive than vapour cell or SQUID-based techniques, the spatial resolution obtained using nitrogen-vacancy centres is unrivalled.



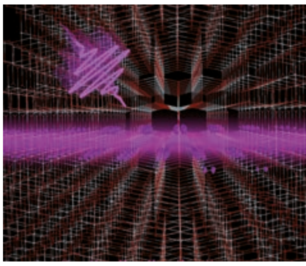
By sweeping the frequency of an applied microwave field and monitoring the fluorescence of the nitrogen-vacancy centres, the optically detected magnetic resonance spectra and thus the magnetic field can be retrieved. Doing this on a pixel by pixel basis and performing a numerical fit to a Gaussian function, a magnetic image for 4 µm diameter magnetic particles can be obtained.

Jānis Šmits

J. Smits et al., Eur. Phys. J. Appl. Phys. 73: 20701, 2016

Light absorption by titanium dioxide (LACUS, Switzerland)

Scientists from Swiss Laserlab-Europe associate partner Lausanne Centre for Ultrafast Science (LACUS) have uncovered hidden properties of titanium dioxide, a promising material for light conversion technology. Using a range of laser-spectroscopic techniques, an international collaboration led by LACUS professor Majed Chergui found that light energy is stored in this material in the form of bound electron-hole pairs (called excitons), confined in two dimensions.



Lattice structure of anatase TiO_2 with a graphical representation of the 2D exciton that is generated by the absorption of light (purple wavy arrow). This 2D exciton is the lowest energy excitation of the material.

Titanium dioxide (TiO_2) appears in different crystalline forms and its 'anatase' polymorph is today one of the most promising materials for a wide range of applications, ranging from photovoltaics and photocatalysis to self-cleaning glasses, and water and air purification. All of these are based on the absorption of light and its subsequent conversion into electrical charges.

The LACUS collaboration has now discovered that the threshold of the optical absorption spectrum is due to a strongly bound exciton, which exhibits two remarkable novel properties: First, it is confined on a two-dimensional (2D) plane of the three-dimensional lattice of the material. This is the first such case ever reported in condensed matter. And secondly, this 2D exciton is immune against temperature and defects as it is present in any type of TiO_2 – single crystals, thin mesoporous films, and even nanoparticles used in devices.

This 'immunity' of the exciton to long-range structural disorder and defects implies that it can store the incoming energy in the form of light and guide it at the nanoscale in a selective way. This promises a huge improvement compared to current technology, in which the absorbed light energy is dissipated as heat to the crystal lattice, making the conventional excitation schemes extremely inefficient. Furthermore, the properties of the newly discovered exciton are very sensitive to a variety of external and internal stimuli in the material (temperature, pressure, excess electron density), paving the way to a powerful, accurate and cheap detection scheme for sensors with an optical readout.

To uncover these peculiar properties of TiO_2, the team used steady-state angle-resolved photoemission spectroscopy (ARPES), which maps the energetics of the electrons along the different axis in the solid; spectroscopic ellipsometry, which determines the anisotropic optical properties of the solid with high accuracy; and ultrafast two-dimensional deep-ultraviolet spectroscopy, used for the first time in the study of materials, along with state-of-the-art first-principles theoretical tools.

Edoardo Baldini and Majed Chergui

E. Baldini et al., Nature Communications 8: 13, 2017

Gold clusters: gentle bond breakers

Synthesis of large carbohydrates is an energy-consuming multi-step process. Therefore, scientists are looking for a catalyst enabling a direct chemical reaction from feedstock molecule methane to larger carbohydrates. Gold nanoclusters seem to be a promising candidate for such a catalyst. In a recent transnational access project performed at FELIX Laboratory (Nijmegen, The Netherlands) an international collaboration consisting of researchers from Radboud University in Nijmegen, Georgia Tech University in Atlanta, US, and Ulm University in Germany, used infrared radiation to show that tiny gold particles can be used to selectively break one single C-H bond in methane. The results of the project were published in the prestigious chemical journal Angewandte Chemie.

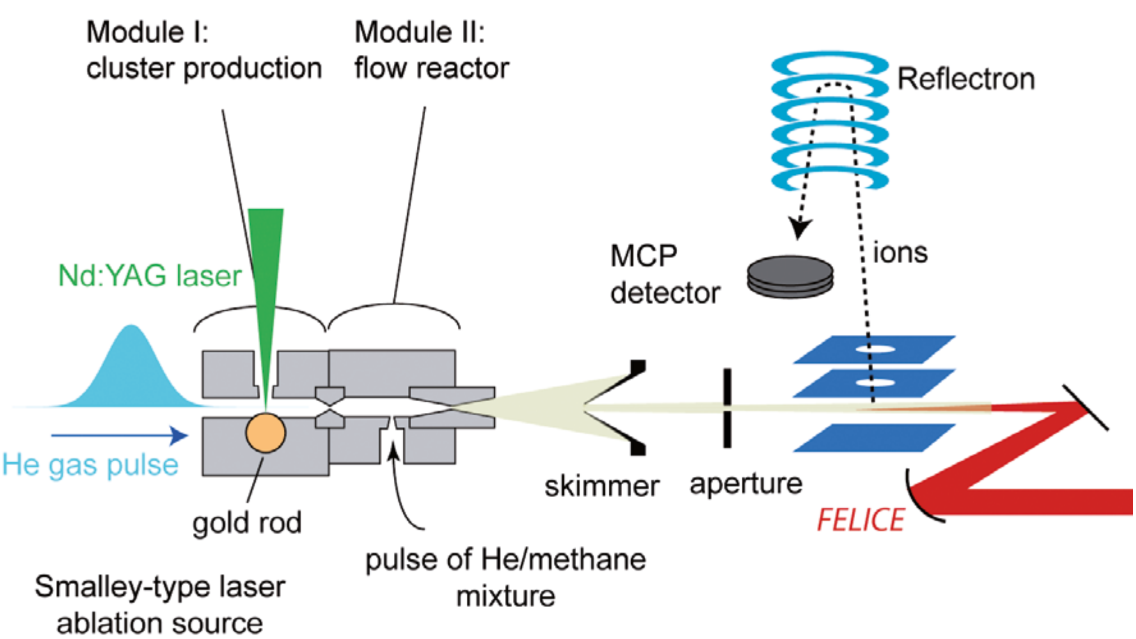
Methane is currently the prime chemical feedstock for larger carbohydrates. The current industrial standard to produce these is to thermally crack methane into smaller molecules and then use these in a mixture called synthesis gas of CO, CO_2 and H_2 to produce the larger carbohydrates, again using plenty of energy. This energy consumption could easily be reduced if a direct chemical reaction from methane (CH_4) to, for instance, ethane (C_2H_6) would be possible. Such a reaction requires a catalyst, which unfortunately has not yet been found.

Catalysts prepare the molecules that need to be chemically transformed into another by weakening certain bonds so that less energy is required for the reaction to succeed. On the other hand, they should not weaken too many bonds at the same time such that the original molecule is modified too much, or even completely destroyed. The working of a catalyst is generally dictated by this fine balance.

Researchers from Radboud University in Nijmegen, The Netherlands, Georgia Tech University in Atlanta, US, and Ulm University in Germany used infrared radiation provided by the FELIX free-electron lasers to find that tiny gold particles possess an important prerequisite for such a catalyst: in linking to the methane molecule, they selectively break one single C-H bond.

Although gold in bulk form reacts very poorly with other substances (the reason why a golden ring never needs polishing) it can become chemically very active when it is reduced to a few-nanometre sized particle, or smaller. Once so small, the quantum nature of the particles starts to become very important, influencing many physical and chemical properties. When these properties change drastically by the addition or elimination of one single atom, the particles are usually referred to as 'clusters'. Gold clusters were famously shown to catalyse the oxidation reaction of carbon monoxide CO, something that requires high temperatures in the absence of a catalyst.

Gold's activity in the catalytic transformation of methane into larger hydrocarbons was known already, too. The partners from Ulm, who specialize in mass-spectrometric experiments measuring the kinetics of cluster-mediated reactions, had found that gold clusters can catalyse the formation of ethane, C_2H_6, from two methane molecules. They, and their colleagues from Georgia Tech, specialists in quantum-chemical theoretical description of such reactions, came up with a reaction mechanism that explained the observed kinetics. There was only one problem: they could not prove the mechanism experimentally, and needed a smoking gun.



The setup at FELIX used for the experiment.

The smoking gun came within reach, when the Ulm researchers ran into a FELIX scientist at an international cluster conference, ISSPIC in Leuven, Belgium. At FELIX, a team have specialised in infrared spectroscopic identification of clusters and their complexes with simple molecules. Over a Tripel Karmeliet, one of Belgium's premium Trappist beers, the idea to identify the structure of one of the gold-methane intermediate products was struck.

The Ulm team made use of the Laserlab-Europe programme for transnational access to FELIX. Here, the joint team produced the gold clusters using laser vaporisation, and had them react with methane. The formed products were irradiated by intense infrared light from the intracavity free-electron laser FELICE, and then analysed using a mass-spectrometer.

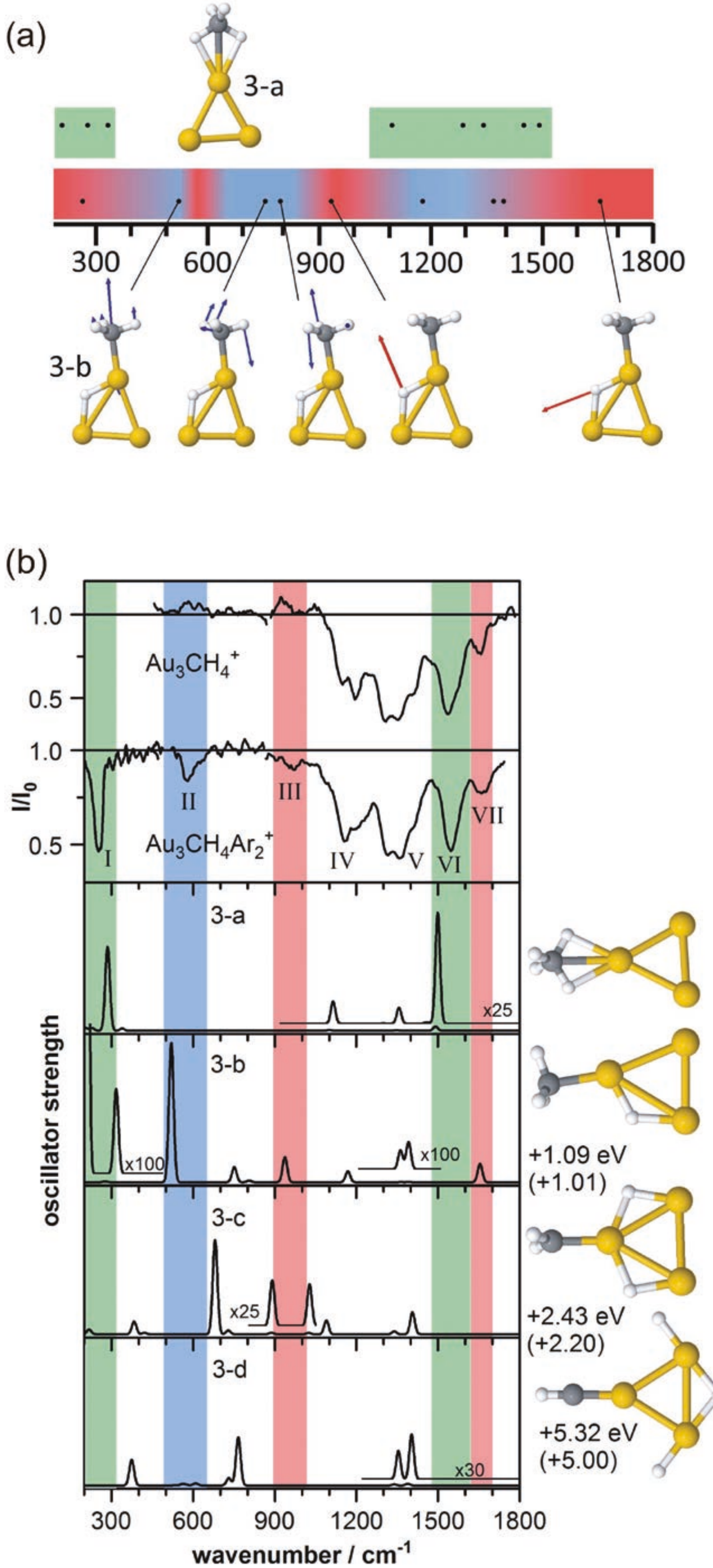
Upon resonant infrared irradiation of a vibrational mode, the gold-methane product fragments, resulting in a depletion of the number of detected products. By recording the number of products as a function of infrared wavelength, a vibrational fingerprint of the product is found.

In these experiments, it was unambiguously demonstrated that methane molecules, when reacting with gold clusters, form a methyl (CH_3) product. For this only one C-H bond of the methane needed to be broken. The use of FELICE was crucial to drive the absorption process for a product that is tightly bound; given the typical photon energies of 0.02-0.1 eV, and binding energies exceeding 2 eV, tens of photons need to be absorbed within a few microseconds.

So why is gold so special? There exist several transition metal elements that easily activate methane and break two bonds, releasing dihydrogen H_2. These elements all have partially filled d-orbitals, which renders them very chemically active. Gold on the other hand has a completely filled d-shell, and should be mostly unreactive, yet it manages to activate methane by breaking just a single C-H bond. This 'gentle' breaking of bonds is attributed to relativistic effects of the gold electronic system, where an s-d hybridisation occurs. In the future design of novel catalysts, this fine balance can be a tuning parameter for the catalyst's activity.

Joost Bakker
FELIX Laboratory (Radboud University Nijmegen)

S. M. Lang et al., Selective C–H Bond Cleavage in Methane by Small Gold Clusters, Angew. Chem. Int. Ed. 56: 13406, 2017



a) General energy ranges for the vibrational modes of gold cluster–methane complexes. The green, blue, and red regions correspond to motions of CH_4, CH_3, and H units, respectively. The black dots correspond to the modes for two different isomers of $Au_3CH_4^+$ and the structures illustrate the nature of the characteristic modes.
b) IR-MPD spectra of $Au_3CH_4^+$ and $Au_3CH_4Ar_2^+$ (upper two panels) as well as calculated vibrational spectra of different structural isomers of $Au_3CH_4^+$. The calculated zero-point energy corrected energies of the isomers (relative to that of the lowest-energy one, isomer 3-a) are given next to their structures. Structural models: Au yellow, C gray, H white.

First step towards a Soft X-ray Laser at the MAX IV Laboratory in Sweden

Stockholm University, Uppsala University, KTH Royal Institute of Technology, Lund University and MAX IV Laboratory in Lund have initiated a project for a Soft X-ray Free Electron Laser at MAX IV. A design study has now received 10 million Swedish krona (1 million euros) from the Knut and Alice Wallenberg Foundation with co-funding by the project partners.

The Soft X-ray Laser (SXL) is an initiative from Swedish users of synchrotron radiation, who together formulated a large set of scientific questions, which only can be addressed by the high power coherent X-rays that a Free Electron Laser (FEL) provides.

The SXL will be based on the already operating 3 GeV linac (linear accelerator) at the MAX IV and will provide high-power pulses of a few femtoseconds in the spectral range of 1-5 nanometres. Pump-probe will be a key user feature, and a number of supporting sources are envisaged.

The conceptual design report of SXL will include detailed theoretical studies of the linac at MAX IV as driver for the Soft X-ray Laser, as well as an analysis of possible improvements to the linac, choices of technology for the undulator section and FEL, and standard configurations of the beamline and its end-stations. The report will be ready in 2020 and then form a basis for future funding.

Swerker Werin (Max IV Laboratory, Lund)

The ELI host countries agree on plan to join the three facilities

The three ELI host countries, the Czech Republic, Hungary and Romania, told the European Commission they plan to merge the three facilities into a single new research consortium in 2018. The plan defines milestones and arrangements for the seat of the new organisation. The announcement, made in Brussels on 1 December 2017, is an important step towards operating the new facility.

Representatives from the countries hosting the Extreme Light Infrastructure (ELI) will merge the three facilities together into a single organisation in 2018. The legal form of the organisation will be a European Research Infrastructure Consortium (ERIC), with the first seat in Romania. The plan is to have the ELI ERIC operating by the beginning of 2019, but many details remain.

The organisation will rotate its legal seat between the countries every three years. Romania will host the first seat, followed by the Czech Republic and Hungary. The move has long been anticipated, but this marks the first time that specific dates and a plan for the seat arrangement have been announced.

In addition to administrative and political support, the European Commission indicated there could be financial support to help the three ELI facilities' transition into a single organisation. That is also based on the fact that the host countries have committed to finance the first two years of operations. The new legal structure of the ERIC allows other member countries time to join as the facilities ramp up to a sustainable operations programme.

Allan Weeks (Associate Director ELI-DC)

Forthcoming events

Network on Experimentation and Best Practices in Biology and Life Sciences Meeting
20 March 2018, San Sebastian, Spain

Network on Extreme Intensity Laser Systems – Annual Meeting
24-25 May 2018, Bordeaux, France

Training Workshop on Time-Resolved Techniques (TReT)
20-22 June 2018, Prague, Czech Republic

To find out more about conferences and events, visit the Laserlab online conference calendar.

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.

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