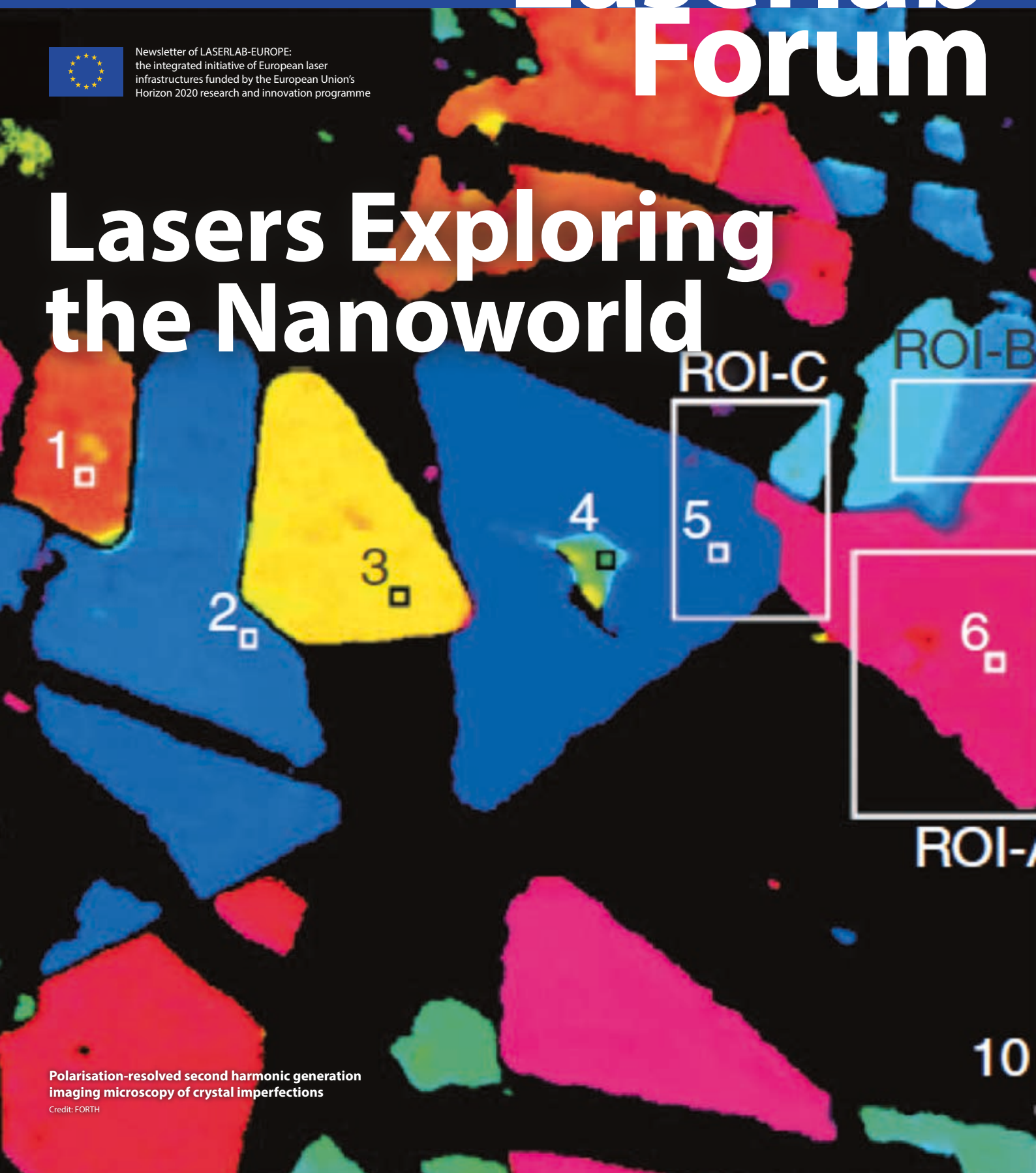


Laserlab Forum



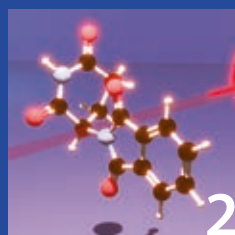
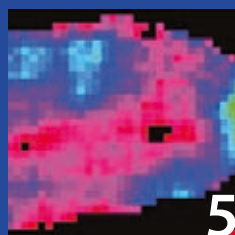
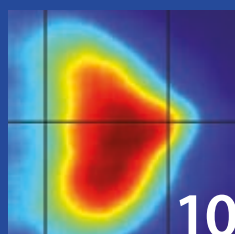
Newsletter of LASERLAB-EUROPE:
the integrated initiative of European laser
infrastructures funded by the European Union's
Horizon 2020 research and innovation programme

Lasers Exploring the Nanoworld



Polarisation-resolved second harmonic generation
imaging microscopy of crystal imperfections

Credit: FORTH

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Editorial



Claes-Göran Wahlström

Laser technology and nanotechnology are two modern and fast developing fields of science, of roughly the same age. The laser was first demonstrated by Theodore Maiman in 1960, stimulated by a 1958 theory paper by Arthur L. Schawlow and Charles H. Townes. In 1959, Richard Feynman gave a legendary talk with the title "There's plenty of room at the bottom", in which he proposed nanotechnology as a new field of research. Since then, the two fields have developed enormously, while mutually supporting and stimulating each other. Lasers are extensively used to both explore and to exploit the rich opportunities of the nanoworld, allowing nanotechnology to develop in a very successful way. Likewise, laser science and laser applications benefit immensely from the many discoveries, inventions and devices resulting from nanotechnology. During the past two decades, when Laserlab-Europe has coordinated joint laser-based research and laser developments, and offered transnational access to external users, research projects bridging between the laser and the nanoscience communities have been frequent and continuously increasing in importance.

"Lasers and the nanoworld" was meant to be the topic of the present issue of the Newsletter, but it quickly became evident that this large topic is better divided into two consecutive issues. The present one gives examples of lasers *exploring* the nanoworld, while lasers *exploiting* the nanoworld will be the topic of the next. I hope you will enjoy them. The present issue further contains, as usual, brief news from several different Laserlab-Europe partner laboratories, with a broad range of research highlights and user access experiments. The COVID pandemic has caused enormous problems, and many tragedies, but it has not stopped science, and it has not stopped Laserlab-Europe from continuing to make progress.

Claes-Göran Wahlström

News

Twisting magnetisation with light – Laser pulses enable faster creation of skyrmions in magnets

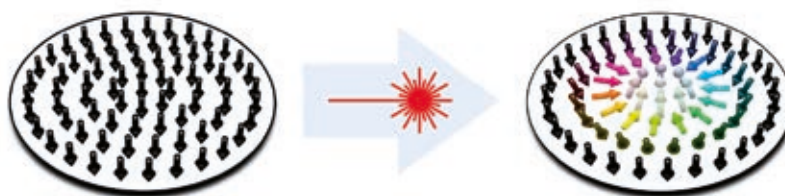
A team of scientists led by MBI and the Massachusetts Institute of Technology has demonstrated how tiny magnetisation patterns (skyrmions) can be "written" into a ferromagnetic material, and have clarified how the topology of the magnetic system changes during this process. The findings are reported in *Nature Materials* (20: 30-37, 2021).

Magnetic skyrmions are tiny "swirls" in the magnetisation of thin films, with the magnetisation pointing in different directions. A single laser pulse of sufficient intensity can create skyrmions with a particular topology – that is,

the magnetisation pattern forms a particular swirl pattern.

The time-evolution of the skyrmion formation was seen by exposing the ferromagnetic film to an optical laser pulse followed by an X-ray laser pulse. The laser pulse excites the system to a state where the magnetisation fluctuates rapidly, allowing tiny "skyrmion nuclei" to form. As the system cools in the presence of a magnetic field, nuclei with a particular swirl pattern grow into the larger skyrmions.

In addition to their usefulness for understanding the basics of topological transitions, since magnetic skyrmions can stably exist at 10 nm diameter and at room temperature, these findings have interesting implications for future concepts of magnetic data processing and storage.

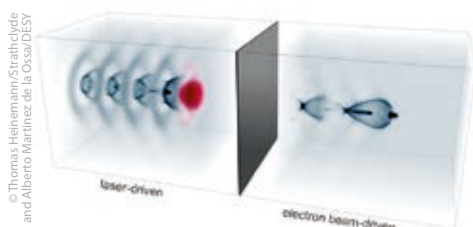


A laser pulse transforms a uniform magnetisation (magnetisation down everywhere) to a skyrmion swirl where the magnetisation in the centre points up.

A new type of miniature plasma particle accelerator

A novel plasma-based particle accelerator has been realised, combining two kinds of plasma accelerators to achieve a rapid energy gain of electrons across a few millimetres. This is the first time this has been achieved outside a very large-scale facility such as CERN or SLAC.

The accelerator takes the intense electron beam produced by laser wakefield acceleration and uses it to drive a separately attached plasma wakefield accelerator. This 'hybrid laser-plasma wakefield accelerator' has been developed in a European collaboration including University of Strathclyde, HZDR, MPQ, DESY and LOA and has passed its first validation tests.



Left panel: Schematic depiction of a laser-driven accelerator (LWFA) with the propagating laser beam shown in red on the left. Right panel: Electrons accelerated by the LWFA are used to drive the second-stage particle accelerator (PWFA).

The accelerator could offer a compact source of high-quality electron beams for applications such as X-ray generation (including X-ray free electron lasers), material science and biomedical research. The work has been published in *Nature Communications* (12: 2895, 2021).

The hybrid plasma accelerator platform and applications can in the future be accessed via SCAPA (the Scottish Centre for the Application of Plasma-based Accelerators) and EPAC (the Extreme Photonics Applications Centre).

APOLLON upgrade well underway

The commissioning of the "short focal length" area of the APOLLON laser facility, operated by LULI, France (<https://apollonlaserfacility.cnrs.fr/en/home/>), using the presently available F2 beam, took place in May 2021 and showcased both the very good characteristics of the laser and the operational capacity of the room. This commissioning took place with laser pulses of 10 J average on-target energy and pulse durations of 25 fs. A range of diagnostics qualified the performance of the facility at this power level. An x-ray image of the zone heated by the laser on the target demonstrated a very good focussing of the laser, with comparable spot sizes as recorded from directly imaging the laser spot size at full power. Solid targets as thin as 2 μm were shot without incurring damage induced by

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.

the pedestal preceding the main pulse, and thus showing very good temporal contrast characteristics of the laser. Emissions of electrons, ions and high energy electromagnetic radiation were recorded, showing good laser-target coupling and an overall performance that is very consistent with what has been reported by similar international facilities.

Surface-sensitive nonlinear XUV-Spectroscopy

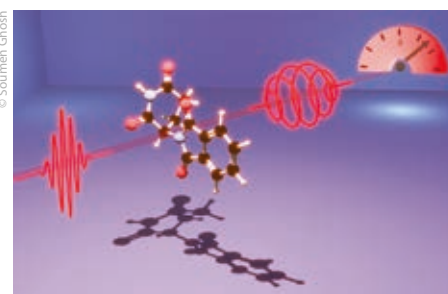
It has so far been difficult to gain a picture of the course of chemical reactions at the atomic level, as this requires measurement methods for extremely short time scales. A collaboration between LOA, the University of California Berkeley and FSU Jena "imported" second harmonic generation (SHG) from the visible regime into the extreme ultraviolet (XUV) spectrum in a table-top setup and have demonstrated the surface specificity of this process. The results have been published in *Science Advances* (7: eabe2265, 2021).

Using the Salle Jaune facilities, XUV radiation was focused down by an ellipsoidal mirror onto an ultrathin titanium foil. The co-propagating incident and emerging SHG beams (at 37.8 and 75.6 eV) were separated by a spectrometer and projected onto a CCD and averaged across several shots. The vast majority of the SHG signal emerges from the incident surface of the titanium foil, showing the surface-specificity of this analysis method. Due to the large number of materials with absorption edges in the XUV-region, this method holds great promise for element-specific measurement in the table-top regime.



3D-sketch of the XUV-SHG setup. Incoming radiation (red) focused on the target and spectral separated by a grating. Spectrum on the camera represents an accumulation of 248 single-shot images.

Molecular chirality in broad daylight



Chirality or the handedness of a molecule plays an important role in biological processes and chemical reactions. Quick and sensitive measurement of chirality, however, is far from being trivial. Now researchers at Politecnico di Milano, Italy have succeeded in developing a highly sensitive optical technique for rapid measurements of chirality across a broad wavelength range using readily available thermal light sources. "Our new technique measures broadband circular dichroism spectra and optical rotation in a few seconds", says Soumen Ghosh, the lead author of the paper published in *ACS Photonics* (8: 2234, 2021). "It allows us to monitor chiral chemical reactions in real-time!"

Laserlab-Europe for a better future – Position Paper published

In a joint position paper, Laserlab-Europe highlights its integrated, cross-domain and multi-faceted approach to address the societal challenges of the Horizon Europe Missions. The paper illustrates the breadth of expertise found amongst the members of the consortium, which covers the whole range of laser science, from fundamental physics to the development of applications addressing diverse global challenges of health, climate and economy. The position paper is available on the Laserlab-Europe website.



ERC Consolidator Grants

The ERC Consolidator Grant is awarded to established researchers with seven to twelve years of experience since completion of their PhD, who present an excellent research proposal and already have a scientific track record showing great promise. The grant is endowed with up to two million euros over a funding period of five years. Among more than 2500 proposals evaluated by the ERC, four proposals from Laserlab-Europe researchers have been successful.

Jochen Mikosch (MBI): Chemistry in motion – Lasers challenge the start-time dilemma



Jochen Mikosch

Jochen Mikosch aims to answer fundamental questions about the structural dynamics of chemical reactions. Efforts to examine the transition states which occur over the course of a chemical reaction have long been hampered by the so-called “start-time dilemma.” Usually, reactants are randomly distributed and oriented in space, and even with an ultra-short laser pulse there is no external control over the precise moment when a reaction takes place. This project addresses these issues by holding the reaction partners closely together in a well-defined initial configuration. The reaction can then be initiated with a femtosecond laser pulse, and by tuning its wavelength the reaction rate can be adjusted. The three-dimensional structure of the transition state is then imaged with a method called Coulomb explosion imaging.

Gerasimos Konstantatos (ICFO): Mid- and long-wave infrared colloidal quantum dot optoelectronics



Gerasimos Konstantatos

Gerasimos Konstantatos will address the high cost and fragmented solutions limiting the potential and wide-spread use of mid-to-long wave IR optoelectronic materials. In order to overcome the fundamental constraints arising from the band-gap of available materials, the team will develop colloidal quantum dot (CQD) technology using intra-band (rather than inter-band) transitions. This will require addressing several fundamental challenges, including developing robust heavy doping schemes and the exploration and control of intra-band relaxation pathways. The advances made in this project will lead to a new disruptive technology for the MWIR/LWIR, as well as providing additional options for other fields that utilise hot carriers, such as catalysis and energy harvesting.

Darrick Chang (ICFO): A new spin on quantum atom-light interactions



Darrick Chang

Darrick Chang aims to demonstrate the enormous and under-exploited potential of interference in light emission for quantum applications. The project aims to establish protocols with exponentially better error bounds than those currently known. The main body of work will be the development of a quantum many-body theory of multiple scattering involving photons and atoms, taking advantage of state-of-the-art tools from condensed matter physics. Beyond robust new routes toward applications, this theory will also reveal exotic new quantum phenomena and lead to new insights into fundamental questions in optics, such as the physical limits of a material's refractive index. Darrick Chang's team anticipate a great advance in the understanding of light-atom interactions and their realm of potential applications.

Leticia Tarruell (ICFO): Unconventional superfluids in quantum gases with competing interactions



Leticia Tarruell

Leticia Tarruell aims to exploit the full potential of ultracold quantum gases with competing interactions, unlocking the observation of unconventional superfluid phases. To this end, the project will explore three distinct mechanisms resulting in unconventional superfluid behaviour: quantum fluctuations, engineered dispersion relations, and interactions with non-zero orbital angular momentum. Exploiting various combinations of bosonic and fermionic potassium atoms will allow the realisation of novel types of ultradilute quantum liquids, supersolid-like gases and liquids, density-dependent artificial gauge fields, and elastic multi-body interactions. The project will also investigate a new approach towards the long-sought px+ipy topological superfluid phase of 2D Fermi gases. These experiments will deepen our understanding of the mechanisms behind unconventional superfluidity, serving the whole range of disciplines where unconventional superfluids or superconductors play a key role.

Lasers Exploring the Nanoworld

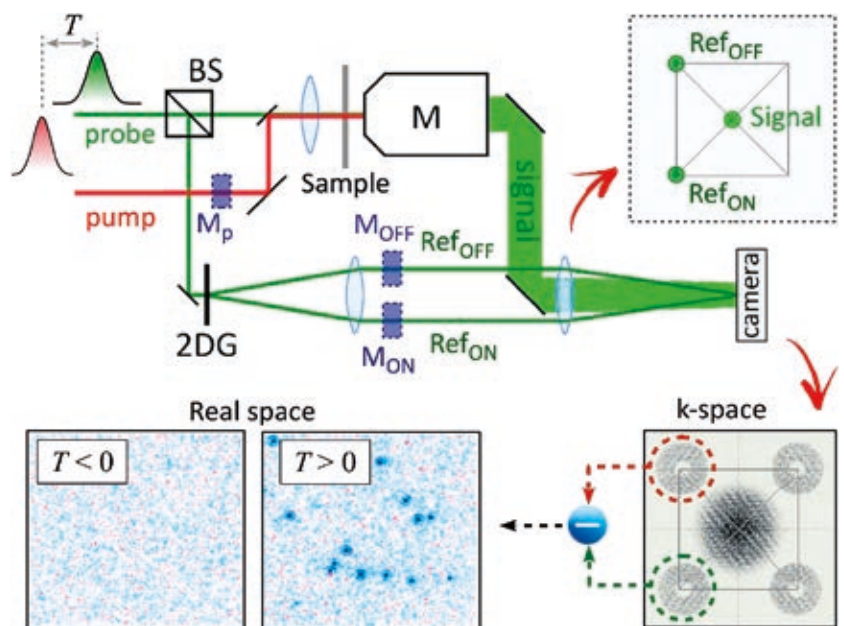
With the advancement of technology over the last half-century, a new frontier has opened up: the nanoscale. Nanometre-scale materials and technologies have the potential to be fast, efficient and effective in unprecedented ways, from metamaterials to specialist chips to lithographic processes. Here we present some of the ways in which Laserlab-Europe's researchers are exploring the science of light and matter at this small scale.

Ultrafast transient holographic microscopy (POLIMI, Italy and ICFO, Spain)

Observing ultrafast light-matter interactions at the nanoscale is of great interest for tracking energy flow in semiconductors or for the study of dynamics in single nanoparticles or even single molecules. This is technically challenging, and most approaches so far have relied on single-point detection, employing raster scanning of a tightly focused beam on the sample to achieve large observation areas. Researchers at ICFO and POLIMI have recently combined ultrashort pulses with off-axis holography to develop a new transient scattering technique that can measure the ultrafast response of hundreds of nano-objects within a wide volume-of-view of around $100 \times 100 \times 100 \mu\text{m}^3$ [1].

The reported approach relies on a dark-field scattering microscope illuminated by 100-fs pulses. While such a microscope can produce wide-field scattering images of the sample in steady state, turning it into a transient microscope usually runs into the problem that cameras have slow frame rates, limiting the ability to modulate a pump beam at high frequencies to achieve sensitive detection. This issue was solved by introducing two off-axis reference beams generated using a 2D grating that interfere with the image at the camera. A 2D Fourier transform of the resulting image pattern neatly separates the interference of the sample image with either reference beam in k-space. By modulating the references synchronously with the pump beam, researchers were able to ensure that one reference interferes with the sample image corresponding to pump-on only, while the other with the pump-off only, effectively de-coupling the pump modulation frequency from the camera acquisition speed and implementing a wide-field digital lock-in. Therefore, each acquired hologram contains both pump-on and pump-off images, which can be isolated in k-space, Fourier transformed back into real space, and subtracted to obtain the transient response at each pixel of the original image.

This concept was demonstrated by acquiring transient scattering signals from dozens of gold nanoparticles scattered on a glass surface in a single measurement and highlighting differences in their ultrafast dynamics due to the particle shape or environment. Furthermore, the holographic setup yields both the amplitude and the phase of the signal, allowing the use of digital holography techniques to propagate out-of-focus signals into focus by a simple data processing step. This was demonstrated by acquiring data with the sample deliberately out of focus. Researchers were nonetheless able to propagate both the



Upper panel: schematic of the multiplexed holographic microscope. BS: beam splitter, M: microscope objective, 2DG: 2D phase grating. Modulators on Ref_{ON} (M_{ON}) and Ref_{OFF} (M_{OFF}), respectively synchronous and asynchronous with the modulated pump beam (M_p), enable recording the pumped and unpumped images, which are separated by k-space filtering. The data show a proof-of-concept wide-field ultrafast transient scattering experiment on a distribution of 100-nm gold nanoparticles.

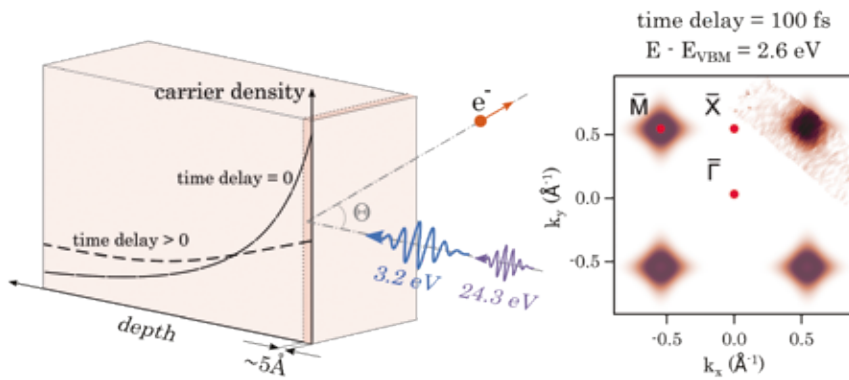
steady state and transient scattering signals into focus, demonstrating the unique 3D capabilities of this approach. It is hoped that this technique can be a single-shot alternative to current photothermal imaging methods.

**Sandro De Silvestri, Giulio Cerullo,
Franco Camargo (POLIMI)**

[1] M. Liebel et al., Nano Lett. 21: 1666-1671, 2021

Ultrafast nanoscale electron transport in solids (LACUS, Switzerland)

When optoelectronic materials with a high absorption coefficient absorb light, strong spatial charge density gradients develop on a nanometer scale at the surface. Consequently, charge carrier transport and recombination processes set in on a femtosecond (fs) timescale to restore equilibrium. Understanding the microscopic details of carrier evolution is a fundamental prerequisite for developing efficient nano-photonics devices. Some applications, such as solar concentrators, light-emitting diodes and lasers, routinely operate under an extremely high charge carrier density. In this regime, complex many-body phenomena start to play an important role in defining the material's



Experimental scheme (left) shows how surface sensitivity of extreme ultraviolet probe pulses allows to study nanoscale charge transport of a solid (here CsPbBr₃) with spatial, momentum, energy and temporal resolution (right).

photophysics, making both the theoretical and technical investigation of their properties difficult. An additional complexity of this ultrafast nanoscale transport stems from its intermediate character between the diffusive and ballistic regimes.

While lateral transport characterisation has seen substantial advances owing to recent developments in imaging and non-linear optics, the surface-to-bulk transport has not been studied to a comparable degree. In the new study carried out at LACUS, time- and angle-resolved photoelectron spectroscopy (TR-ARPES) with extreme ultraviolet (EUV, 15–50 eV) probe and ultraviolet (3.2 eV) pump pulses was applied on single crystals of the CsPbBr₃ perovskite. The experiment is schematised in the figure. By exploiting the exceptional surface sensitivity of photoelectrons emitted by EUV photons, obtained at the Harmonium light source, the surface electronic population was followed in reciprocal space, providing a sub-nanometric picture of the light-induced dynamics with fs temporal resolution.

The experimental findings, supported by theoretical modelling of nanoscale carrier diffusion and recombination under strong optical excitation, reveal the super-diffusivity of charge carriers, explainable by the high band velocities of hot carriers within a quasi-ballistic transport regime. The transport was found to asymptotically approach a conventional diffusive regime at the highest carrier densities even at sub-ps timescales, due to an onset of strong electron-hole scattering, limiting the carrier mean free path. The results showcase the capability of EUV-based TR-ARPES to study ultrafast nanoscale transport with momentum selectivity in crystalline solids.

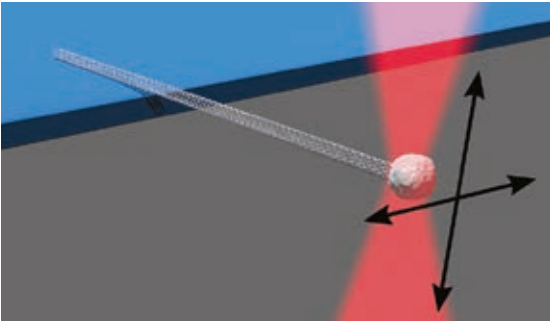
Majed Chergui (LACUS)

Exploring the stiffness of nano-objects (ICFO, Spain)

The vibrations of the atoms within a solid material are determined by their temperature. When the temperature is higher, the intensity or amplitude of the vibrations is larger. These vibrations directly affect the stiffness of the solid material. Even though scientists are aware of the fact that there is a temperature-dependent change of the elasticity in a single nanoscale system, so far, it still had not been experimentally detected.

In a study published recently in *Physical Review Letters*, ICFO researchers, in collaboration with the Instituto de Nanociencia y Materiales de Aragón (INMA) of the University of Zaragoza, ICMM-CSIC, Polytechnic University of Marche, TU Delft, and the University of Nottingham, have reported on a new approach to measure the small change in the elasticity of a nanotube when changing its temperature.

In their experiment, the team built a 1-10 micrometre long carbon nanotube, called a cantilever, where one of its ends was attached, fixed to a silicon chip, and the other end was free to bounce around in the air. Next, on the free end of the nanotube, the scientists deposited a tiny amount of platinum to form a particle. Then they placed the entire system within a chamber at room temperature, and while lowering the temperature slowly down to a few degrees Kelvin, they illuminated the nanotube with a He-Ne laser and observed how the system vibrated. They found and measured the vibration mode with lowest frequency, knowing that its motion amplitude was the largest. They observed how the resonance frequency changed to measure the stiffness of the nanotube, which is quantified by the so-called Young's modulus.



Schematic illustration of the mechanical resonator with the platinum particle at the end of the cantilever.

Now, the energy dissipation of resonators is described by the thermal bath, or thermal reservoir, which describes how the resonator is coupled to the outside world. So far, it had been very difficult to identify the microscopic nature of the thermal bath, but in this study, the team was able to show that the thermal reservoir is composed of phonons by a sizeable amount. While lowering the temperature, they saw that the stiffness of the nanotube was dependent on these phonons, that is, the collective excitation in a periodic, elastic arrangement of atoms within the solid.

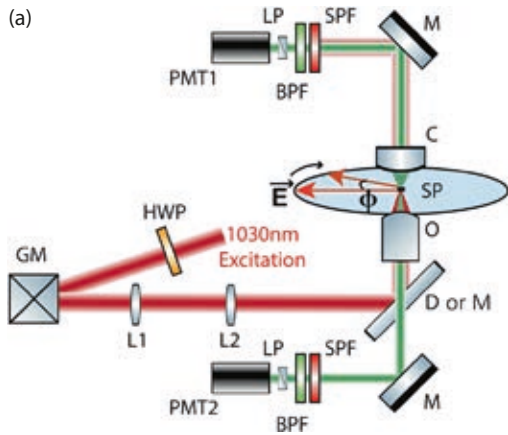
These measurements enable a better comprehension of the physics of mechanical resonators and pave the way to optimising them to reach unprecedented levels of sensing precision.

Adrian Bachtold (ICFO)

S. Tepsic et al., *Phys. Rev. Lett.* 126: 175502, 2021

Polarisation-resolved second harmonic generation imaging microscopy of 2D materials (FORTH, Greece)

The emerging family of graphene and related two-dimensional (2D) materials has provided researchers with fertile ground for exploring fundamental physical phenomena and developing innovative technological solutions. 2D transition metal dichalcogenides (TMDs) MX₂ (M: Mo or



W, and X: S, Se, or Te) are direct-bandgap semiconductors (structurally similar to graphene), that show electronic and optoelectronic properties, along with desirable mechanical strength and flexibility, offering great promise for use in future electronic devices.

Lately, nonlinear optical measurements, particularly second harmonic generation (SHG) used in conjunction with laser raster-scanning microscopy, have created new opportunities for improving the image resolution of 2D crystals [1, 2] (Figure a). In particular, the SHG signal depends on the elements of the second-order susceptibility tensor $\chi^{(2)}$, which are non-vanishing only for non-centrosymmetric media such as the atomically thin TMDs. At the same time, the polarisation of the SHG field depends crucially on the 2D crystal symmetry and orientation (Figure b).

Based on such SHG signal dependencies, the crystal quality of TMDs can be evaluated using polarisation-resolved SHG (P-SHG) imaging, with resolution which is limited only by the optical limit of diffraction [1, 2] (Figure c).

Moreover, atomically thin TMDs can be assembled in vertical stacks held together by van der Waals forces, enabling interlayer coupling between the layers. This creates new physical properties that depend on the relative orientation (twist angle) between the TMD monolayers. P-SHG imaging microscopy provides accurate and real-time measurement of the twist angle, which is of utmost importance for characterising a 2D TMD heterostructure [3] (Figure d).

Further, degenerate minima in momentum space valleys in 2D materials provide an additional degree of freedom that can be used for information transport and storage. When these atomically thin crystals interact with intense laser light, the SHG field shows special characteristics that reflect not only the broken inversion symmetry in real space, but also the valley anisotropy in reciprocal space. This anisotropy is present whenever there exists a population imbalance between the two valleys (VPI). SHG intensity is dependent on the excitation field polarisation. The variation of this dependence with temperature, as revealed by P-SHG imaging, is a unique fingerprint of VPI [4] (Figure e).

Optical P-SHG imaging is envisaged as a powerful tool for the characterisation of 2D TMD heterostructures, and for the engineering of their physical properties for emerging applications.

**Sotirios Psilodimitrakopoulos and
Emmanuel Stratakis (FORTH)**

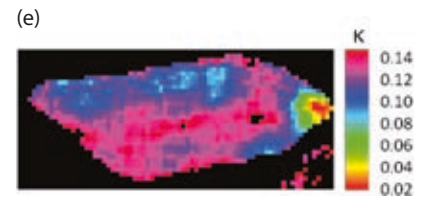
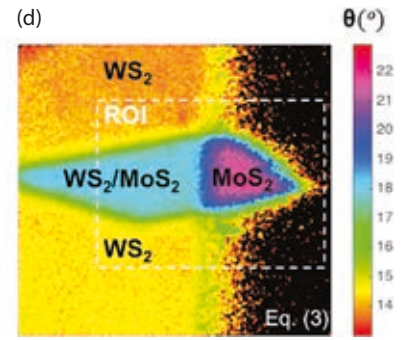
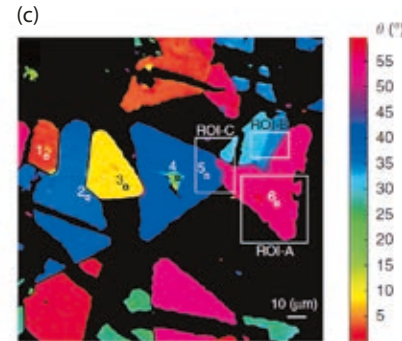
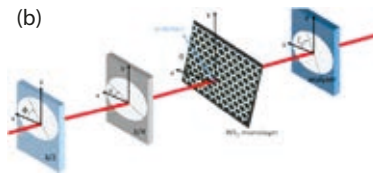
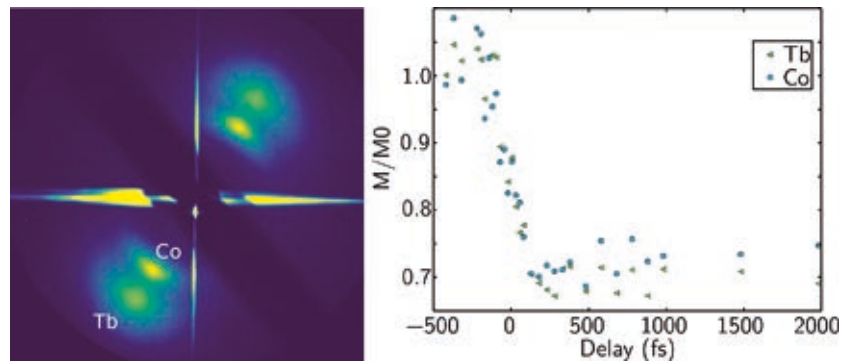


Figure (a) Experimental setup for P-SHG raster-scanning imaging microscopy. (b) Coordinate system for the theoretical model describing the P-SHG signals from 2D TMDs. (c) P-SHG imaging of crystal imperfections. (d) Real-time imaging of twist-angle. (e) P-SHG imaging of VPI.

Femtosecond magnetisation dynamics for faster magnetic storage (LOA, France)

Our society's ever-growing need for computing power requires the regular development of new electronic devices. In the domain of magnetic recording, this translates into faster systems with higher capacities. To go beyond the present state of the art, a deep understanding of femtosecond magnetisation dynamics at the nanometre length scale is necessary.

The Sources for Interaction, Imaging & Medical application group (SIIM) is studying femtosecond magnetisation dynamics in rare earth transition metal alloys, which are promising materials for faster magnetic recording devices [1]. In these materials, the magnetisation dynamic of each component needs to be followed independently. Here, a $\text{Co}_{88}\text{Tb}_{12}$ alloy was observed, which exhibits nanoscale magnetic domains. Such a magnetic structure acts as a diffraction grating for the wavelengths corresponding to the absorption bands of Co and Tb. Using the high harmonic source of the Laboratoire d'Optique Appliquée (LOA), fem-



Magnetic scattering pattern of a CoTb alloy film obtained at photon energies between 40 and 70 eV with the high harmonic source at LOA, showing 2 peaks corresponding to the Co M2,3 bands and Tb O1 band. Following the intensity of these two edges allow us to follow the magnetisation dynamics of Tb and Co independently on the femtosecond time scale.

[1] S. Psilodimitrakopoulos et al., Light Sci. Appl. 7: 18005, 2018
[2] G. M. Maragkakis et al., Opto-Electron. Adv. 2: 190026, 2019
[3] S. Psilodimitrakopoulos et al., 2D Materials 8: 015015, 2021
[4] L. Mouchliadis et al., npj 2D Mater. Appl. 5: 6, 2021

to second pulses can be generated spanning the photon energy range of 40 to 70 eV, covering the M_{2,3} band of Co (around 60 eV) and the O₁ band of Tb (around 45 eV). This leads to the appearance of two different scattering spots on a CCD camera set a few centimetres downstream of the sample (see figure, left).

LOA researchers have recently studied similar samples at the Free Electron Laser FERMI (Trieste, Italy) and were able to follow for the first time the ultrafast dynamics of the magnetic anisotropy in these alloys, a key parameter that must be precisely controlled for the realisation of magnetic recording devices [2]. Comparing to this study, the work at LOA allows to simultaneously look at the photons in resonance with the Co and Tb bands thanks to the large bandwidth of the high harmonic source. In this particular case, very similar magnetisation dynamics for Co and Tb are observed, contrary to the previous investigations (see figure, right). This on-going work will allow to fine-tune the composition of the alloy and optimise its properties for particular applications.

Boris Vodungbo (Laboratoire de Chimie Physique-Matière et Rayonnement) and Guillaume Lambert (LOA)

[1] C. D. Stanciu et al., Phys. Rev. Lett. 99: 047601, 2007

[2] M. Hennes, Phys. Rev. B 102: 174437, 2020

Transient X-ray grating spectroscopy for nanoscale measurements (LACUS, Switzerland, LENS and FERMI, Italy)

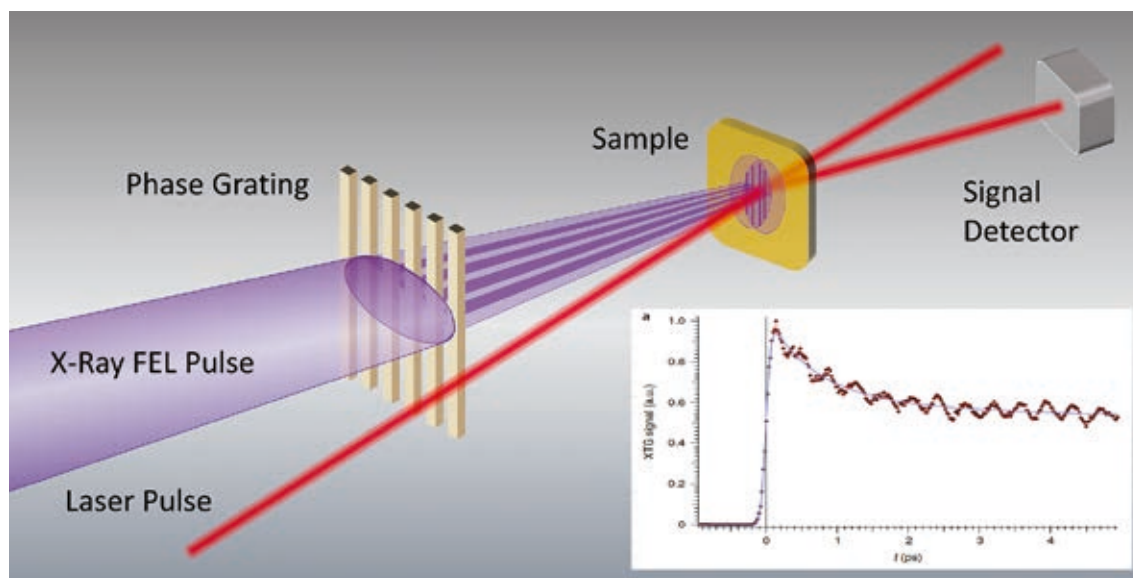
Measuring a variety of transport phenomena at the nanoscale is becoming vital with the miniaturisation of devices. Yet it remains elusive due to the lack of methods which are precise at such length scales. Transient grating (TG) spectroscopy is the traditional tool for measuring transport phenomena. It uses two laser pulses to activate a medium by creating an interference pattern, or grat-

ing, from stripes of excitations that can be thermal, electronic, magnetic or even structural. The modulation depth of the grating can be measured by using it to diffract a third, time-delayed probe beam, which monitors the grating evolution as it fades away due as the initial excitation propagates through the material. The grating spacing is determined by the wavelength of the laser pulses used to create it and is on the order of hundreds of nanometres to microns in the visible-ultraviolet range.

Transport properties at the nanoscale are expected to greatly differ from those at the meso- and larger scales. In particular, a change of regime from ballistic to diffusive is known to occur, yet it has not yet been unambiguously observed. Harnessing such nanoscale transport phenomena calls for the use of short-wavelength radiation and, in particular, X-rays. However, crossing two X-ray beams in order to generate a grating with nanometre step sizes is a challenge. In an international collaborative effort led by the Paul-Scherrer-Institut (PSI), Switzerland, and involving several Laserlab-Europe partners (LACUS, LENS and FERMI), the Talbot effect (see figure) was exploited to create such gratings using beams of 0.17 nm wavelength at the Swiss Free-Electron Laser (SwissFEL, PSI). The evolution of the generated grating was probed by a time-delayed 400 nm probe pulse. The results, revealing the optical and acoustic response in the form of coherent phonon oscillations over hundreds of femtoseconds to tens of picoseconds (Figure inset), appeared in *Nature Photonics*. This first demonstration of hard X-ray transient grating spectroscopy opens the way to novel and exciting developments in the study of nanoscale transport phenomena. The next step will be to replace the 400 nm probe pulse by a hard X-ray probe pulse, thus permitting access to the nano length-scale directly.

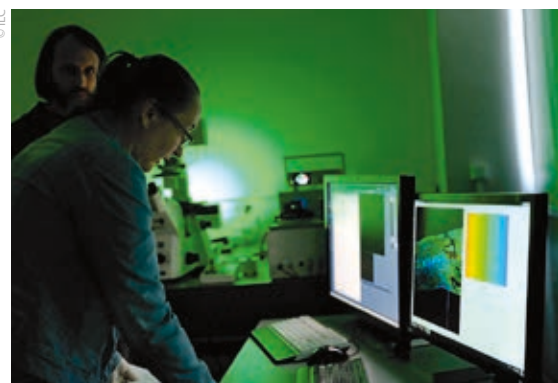
J. R. Rouxel and M. Chergui (LACUS), S. Catalini and R. Torre (LENS), D. Fainozzi and C. Masciovecchio (FERMI), R. Mankowsky and C. Svetina (PSI)

J. R. Rouxel et al., Nat. Photonics 15: 499-503, 2021



X-ray TG setup scheme: The XFEL pulse (in purple) is diffracted by a transmission phase grating, generating a Talbot carpet on the sample, which induces a TG excitation with spatial periodicity. A delayed optical pulse (in red) monitors the temporal evolution of the XRG via transient diffraction. In the inset, XRG signal from a bismuth germanate sample at 7.1 keV with an excitation grating pitch of 770 nm. The fast oscillations are attributed to a coherent optical phonon.

User training in the time of coronavirus: first online workshop on Data Analysis in Time-Resolved Imaging and Spectroscopy



Hands-on session on FLIM data analysis

The User-Training Workshop on Data Analysis in Time-Resolved Imaging and Spectroscopy (TRIS) was organised in May 2021 by the International Laser Centre of CSTI SR (ILC), Bratislava, in the framework of the Laserlab-Europe User Training schools. The main aims of the workshop were to attract new users of Laserlab Europe services, to train a new generation of researchers to employ time-resolved technologies in their professional studies, and to exchange knowledge and initiate collaborations among the participants.

The one-day workshop consisted of seven online lectures, followed by sessions dedicated to hands-on training. It put together an interesting international group of almost 20 students, scientists and professionals. The first set of lectures provided participants with a basic introduction to modern time-resolved detection techniques and their applications in spectroscopy and imaging. The second block of lectures was focused on applications such as fluorescence lifetime imaging (FLIM), time-resolved image processing and time- and angle-resolved photoelectron spectroscopy. The lectures were followed by three successive hands-on sessions, each for a different set of participants, which were dedicated to practical analysis of complex data such as FLIM of cells, tissues, and whole animals (fireflies), chlorophyll autofluorescence of plants and algae, and NAD(P)H spectroscopy in live cells and various solvents.

Thanks to the Laserlab-Europe support, ILC utilised the opportunity to gather experts and professionals in the field of laser physics, time-resolved detection and biomedical research from leading European infrastructures. The broad range of expertise present in Laserlab-Europe's networks was key to successfully implement the workshop, which was highly appreciated by its participants.

Dusan Chorvat (ILC)

Lasers Fighting Cancer Symposium

A virtual academic-industrial symposium on exploring the use of lasers in combating cancer was held on 25 May 2021. The aim of this Laserlab-Europe event was to raise awareness of the opportunities for laser technologies, to inform about barriers that new technologies face when being deployed in the medical/pharmaceutical sector and what needs to be done to overcome them, to showcase Laserlab-Europe activities and expertise to the broader community, and to initiate discussion on joint R&D activities.

The half-day event brought together international academic researchers and medical professionals, as well as an industrial perspective from companies engaged in technology commercialisation. Topics included clinical translation of laser spectroscopy, vibrational imaging approaches, radiotherapy with very high energy electron beams and the use of optical systems for an improved cancer diagnosis and therapy. The talks showcased the role lasers play from understanding cancer to advanced diagnostics up to

treatment modalities – on the level of fundamental or pre-clinical research up to the clinical application. In addition, the huge global market for medical applications, e.g. for optical and radiological medical imaging technologies, minimal-invasive individualised treatment techniques and optical drug monitoring, was highlighted.

The online forum enabled more than 140 participants and speakers to attend from across the European cancer community as well as from the US. The event was well-received, and targeted follow-up events are planned for the future.



Photodynamic therapy of a patient with head and neck cancer.

Radiation properties of high-power laser generated supersonic jets and shocks

The experimental campaign performed at the Laserlab-Europe partner PALS was dedicated to the study of supersonic plasma jets and shocks, with particular focus on the radiative properties of their formation and evolution. The findings showed that the density and topology of the ambient plasma had a significant effect on the shock geometry, both by directly altering the plasma parameters in the interaction region, but also indirectly by its influence on the jet formation. The latter highlights the importance of understanding the impact of the initial jet geometry on the observed shock dynamics. One of the goals of the work was also to shed light on the interaction of jets and shocks occurring in various astrophysical phenomena such as Herbig-Haro objects and accretion discs. This experiment was led by scientists from ELI Beamlines in collaboration with Helmholtz-Zentrum Dresden-Rossendorf, Technische Universität Dresden, Czech Technical University, Institute of Physics and Institute of Plasma Physics of the Czech Academy of Sciences, CELIA at University of Bordeaux, and Institute of Applied Physics and Computational Mathematics and Xi'an Jiaotong University in China.

In contrast to commonly used methods for producing plasma jets, such as utilising complex target geometries, the annular intensity profile of the PALS laser allows for jet generation using a defocused beam onto a flat target. The third harmonic frequency of the PALS laser (at a wavelength of 438 nm), with an energy in the range of 70 – 110 J was used in the experiment, and the corresponding intensity on target was $I_L \leq 10^{14}$ W/cm² for the focal spot radius of $r_L = 300\mu\text{m}$. The shocks were formed through the subsequent collisional interaction of the jet with a background argon plasma produced with a de Laval nozzle placed a few millimetres in front of the copper target.

The plasma jet and shock parameters were characterised using several diagnostics working both in the optical and X-ray spectral range. The formation and evolution of the jet and shock was studied with a three-frame optical

imaging system developed by the Institute of Plasma Physics and Laser Microfusion in Warsaw. Shadowgraphy was used to study the shock structures, and interferometry for measurements of the electron density. A four-frame X-ray pinhole camera developed at Czech Technical University, together with an X-ray streak camera, also allowed for the characterisation of the temporal evolution of the X-ray emission produced by the jet and shock.

Shadowgraphy of a typical jet is shown in Figure 1a. The jets in the experiment had an electron density of $n_e \sim 10^{19}$ cm⁻³, a radius of approximately 0.1 – 0.15 mm, and extended several millimetres from the target. The jet velocity was obtained from the shadowgraphy and X-ray streak camera measurements, and was in the range of 400 – 700 km/s depending on the laser energy. Figure 1b shows shadowgraphy of a shock formed through the interaction

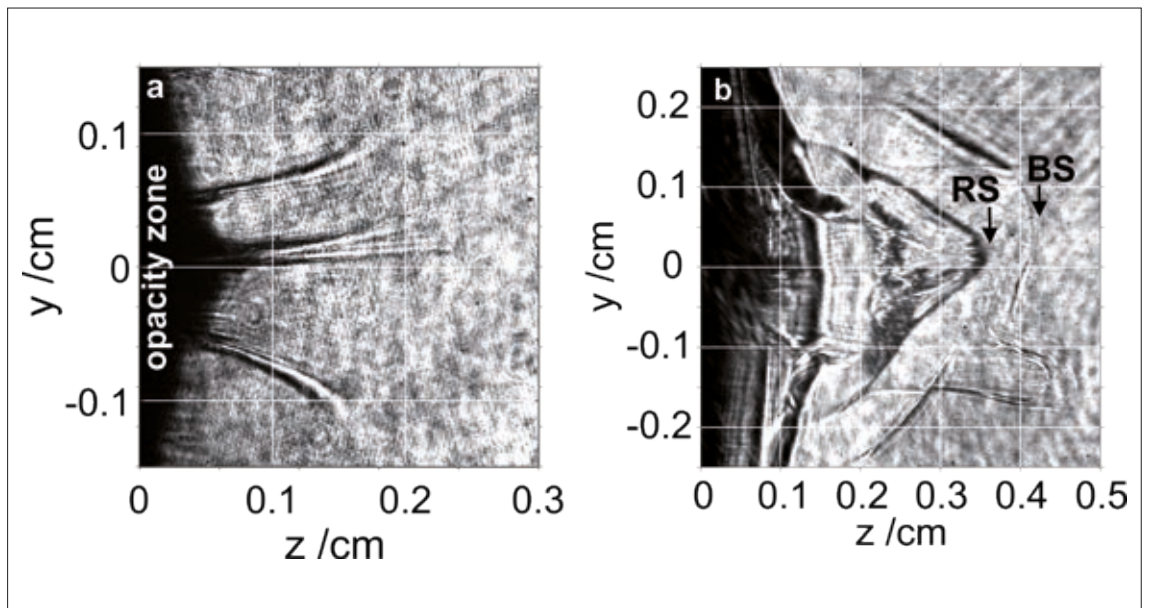


Figure 1: a) Shadowgraphy of a typical jet created by the interaction of the laser with a copper target (at $t = 6.5$ ns, and for a laser energy of $E_L = 71$ J). b) Shadowgraphy of a typical shock formed through the interaction of the jet with the argon background plasma (at $t = 8$ ns, $E_L = 110$ J, and argon neutral density $n_{Ar} \sim 4 \times 10^{18}$ cm⁻³). The arrows indicate the bow shock (BS) in the argon, and the tip of the reverse shock (RS), which slows the jet and results in an accumulation of copper plasma downstream.

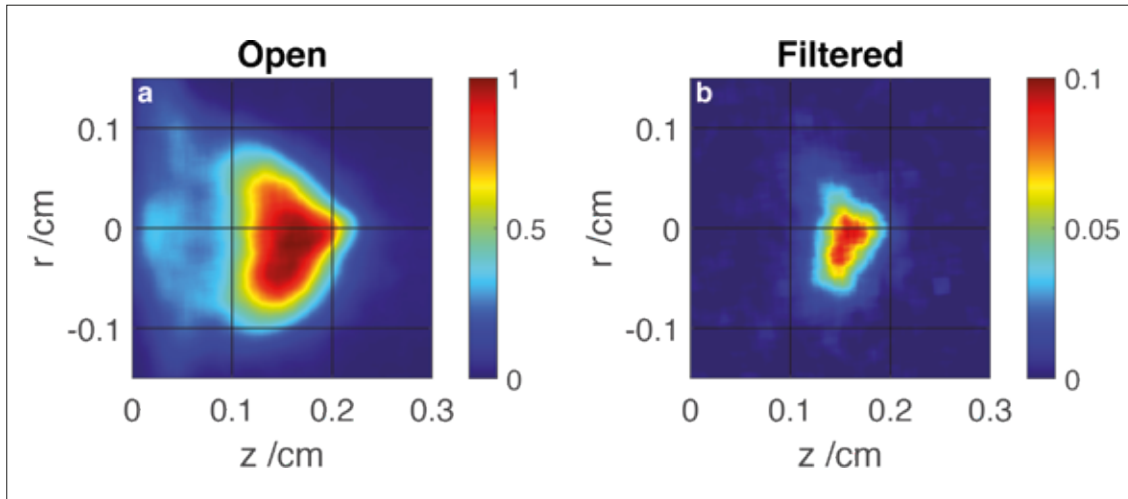


Figure 2: X-ray emission in the photon energy range of $E_{ph} = 10 - 1000$ eV captured with the pinhole camera. a) Open and b) filtered X-ray emission from the shock ($t = 5$ ns, $E_L = 85$ J, and $n_{Ar} \sim 4 \times 10^{18}$ cm $^{-3}$).

of the copper jet with a background argon plasma. A clear bow shock is visible in the argon plasma, with interferometry measurements indicating a factor of two higher density compared to the background plasma. A strong reverse shock is also seen in the copper plasma, with an order of magnitude increase in density due to the deceleration of the jet and accumulation of material. The shock geometry was found to be highly dependent on the background density through its influence on the jet structure and formation. The gas was seen to inhibit the radial plasma expansion in the target region, leading to a redirection of the flow and collimation of the jet.

The shock structure and its radiation characteristics were studied further using an X-ray pinhole camera, and an example is shown in Figure 2. Comparison with the optical imaging diagnostic showed the strongest X-ray emission from the copper plasma region. The use of filtered imaging, and comparison with the spectral intensity obtained from atomic models, also allowed for the electron temperature to be determined. It was found to be in the range of $T_e = 50 - 150$ eV for the jet, and around $T_e \leq 150$ eV in the shock heated copper.

Supersonic jets, and shock formation through their interaction with ambient plasma, are ubiquitous in astrophysics. While the vastly different time and length scales mean they cannot be directly reproduced in the laboratory, dedicated experiments allow for a detailed study of the underlying fundamental mechanisms driving them. Furthermore, comparison of relevant dimensionless parameters such as the Mach, Peclet and Reynolds numbers, and the jet-to-ambient density ratio, allows for a scaling of the laboratory observations to astrophysical phenomena that exhibit similar hydrodynamic behaviour. Based on the parameters obtained in the experiment, the observations can provide insights into the formation and evolution of Herbig-Haro objects, which are associated with jets and shock phenomena often occurring in star forming regions. The dimensional analysis indicates that experimental time-scale on the order of 10 ns roughly correspond to hundreds of years for the Herbig-Haro objects.

Hannes Bohlin (ELI Beamlines)

H. Bohlin et al., Plasma Phys. Control. Fusion 63: 045026, 2021

ELI's establishment as an ERIC and start of operations

© ELI Beamlines



ELI Beamlines in Dolní Břežany, Czech Republic

On 30 April 2021, the Extreme Light Infrastructure (ELI), one of Laserlab-Europe's close collaborators, was granted the legal status of European Research Infrastructure Consortium (ERIC) by the European Commission. The establishment of ELI ERIC opens the door for researchers, industry and countries to gain access to the world's largest collection of super powerful and ultra-fast lasers for science and enable cutting-edge research in physical, chemical, materials, and medical sciences, as well as breakthrough technological innovations.

The Czech Republic hosts the ELI ERIC statutory seat in Dolní Břežany at the ELI Beamlines facility. A second facility, ELI-ALPS, is hosted by Hungary in Szeged. Italy and Lithuania also joined as founding members, while Germany and Bulgaria are founding observers. A third

ELI facility is under construction in Romania in the field of nuclear photonics and is expected to complement the current ELI ERIC facilities in the future.

The integration of the ELI facilities into a single organisation is planned to take place over the next two to three years. During that time, management, technical and scientific procedures in the different facilities are being harmonised. The merger is being facilitated by a 20 million euro grant, called "IMPULSE", from the European Union under the Horizon 2020 programme.

ELI ERIC has officially started operations with the first General Assembly meeting on 16th June 2021 resulting in the appointment of Allen Weeks as Director General of ELI ERIC and Caterina Petrillo as Chair of the ELI ERIC General Assembly.



ELI-ALPS in Szeged, Hungary



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 technical feasibility and 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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Laserlab-Europe AISBL Expert Working Groups

With the goal to boost collective action among its members and to strengthen the quality and impact of the research carried out at each facility, Laserlab-Europe AISBL is establishing Expert Working Groups for topics of strong common interest. The first four Expert Groups address the following areas:

- Laser science for cancer research
- Lasers for clean energy
- Laser-generated electromagnetic pulses
- Micro- and nano-structured materials for experiments with high-power lasers

Kick-off meetings of the Expert Working Groups have been held earlier this year, provid-

ing a platform for all participants to showcase their research in the respective field. External interested collaborators bring in additional expertise so that the groups cover a broad range of research and applications. Together, the groups are developing strategies for new collaborations and joint approaches to complex problems. Brief descriptions can be found at www.laserlab-europe.eu/aisbl/expert-groups.

Laserlab-Europe AISBL is established as a not-for-profit organisation. It currently has 45 members from 22 countries and is open to new members and new joint activities triggered by the participants.