LASERLAB USER MEETING

"UPDATING OPTICAL AND LASER METHODS FOR ENERGETICS, MATERIALS, CHEMISTRY AND BIOMEDICINE "

28-29 March 2011

Istituto Nazionale di Ottica

CNR Campus - Pisa, Italy

Book of abstracts

Foreword by the Meeting Chairs

LASERLAB EUROPE is an Integrated Initiative of European Laser Research Infrastructures. Presently, in the second phase of its successful cooperation, it is constituted by a Consortium of 26 Laser Research Infrastructures from 16 European member states. One of the most important objectives of the Consortium is to offer Access to European researchers in a coordinated fashion through a dedicated Transnational Access Programme.

Annual User Meetings are organized at Consortium level to bring together a significant number of Users, facilitating and fostering their exchanges with the infrastructures. These Meetings also serve for the Access providers to present to the User community recent upgrades, new experimental set-ups or diagnostics, and to the Users to disseminate their scientific results.

The present Meeting, under the topic **"UPDATING OPTICAL AND LASER METHODS FOR ENERGETICS, MATERIALS, CHEMISTRY AND BIOMEDICINE"**, has been organized by the Board of User Representatives of LASERLAB Europe, and is chaired in this occasion by Marta Castillejo and Antonio Giulietti. The Meeting includes 3 keynote invited talks, given by internationally renowned scientists, 23 contributions (oral and posters) and a closing Round Table where issues of concern to Users will be discussed.

We are confident that the Program is of interest to attendees and to the whole LASERLAB community. We would like to express our gratitude to LASERLAB Facilities for suggesting speakers, to the presenters who have accepted to participate at short notice, to Istituto Nazionale di Ottica, CNR, and its Pisa Unit, for kindly hosting the Venue, to the Board of User Representatives for their organizational support, and specially to Daniela Stozno and her team for providing efficient help during preparations of the Meeting. We also thank Petra Koester (INO) for acting as Editor of this Book of Abstract.

Marta Castillejo

Antonio Giulietti

User Representatives of LASERLAB Europe

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Chairs

Marta Castillejo, Instituto de Química Física Rocasolano, CSIC, Madrid, Spain

Antonio Giulietti, Istituto Nazionale di Ottica, CNR

Unit "Adriano Gozzini", Pisa, Italy

MONDAY, MARCH 28th

08:30-12:00 **Registration**

08:45-09:00 **Opening address by INO Director and Meeting Chairs**

09:00-09:20 **Presentation of the transnational Access Program of LASERLAB-Europe II,** Pascal d' Oliveira, SLIC, Paris, France

Session 1. Interactions at Extreme Energy Density

Chair: Antonio Giulietti, INO, Pisa, Italy

09:20-09:50 **Keynote presentation: The collaboration of European FEL facilities**, Josef Feldhaus, Deutsches Elektronen-Synchrotron, DESY, Hamburg, Germany

09:50-10:10 **Optical field ionized XUV laser using the guiding of the pump beam in a plasma performed channel,** Jaroslav Nejdl, Czech Technical University in Prague, Prague, Czech Republic

10:10-10:30 **Electron beams and X-ray radiation generated by laser wakefield in capillary tubes**, Brigitte Cros, Université Paris Sud, Orsay France

10:30-10:50 **Measurement of harmonics produced in gas-puff laser plasmas by synthetic single-crystal diamond detectors,** Claudio Verona, Rome University Tor Vergata, Rome, Italy

10:50-11:20 **COFFEE BREAK**

11:20-11:40 **Proton acceleration and x-ray emission in fast electron transport studies**, Leonida Antonio Gizzi, INO, Pisa, Italy

11:40-12:00 **Radiative Shocks at PALS**, Chantal Stehle, Observatoire de Paris, Meudon, France

12:00-12:20 **~2.1 µm laser based on Ho:KLu$(WO_4)_2$ crystal in-band pumped by a Tm:KLu$(WO_4)_2$ laser and a diode laser,** Xavier Mateos, Universitat Rovira i Virgili, Tarragona, Spain

12:20-12:40 **Three-dimensional photonic quasicrystals,** Georg von Freyman, Institute of Nanotechnology, Karlsruhe and Department of Physics, Kaiserslautern, Germany

MONDAY, MARCH 28th

12:40-15:00 **LUNCH AND POSTER SESSION**

Poster 1 **Influence of Laser-Plasma Parametric Instabilities in Shock Ignition Relevant Regime**, Carlo A. Cecchetti, INO-CNR, Pisa, Italy

Poster 2 **The Laserlab ACCESS program at PHELIX**, Bernhard Zielbauer, GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany

Session 2. Ultrafast Laser Probing of Matter

Chair: Marta Castillejo, CSIC, Madrid, Spain

15:00-15:30 **Keynote presentation: Probing primary photoinduced processes in organic molecules with tunable few-optical-cycle light pulses**, Giulio Cerullo, Politecnico di Milano, Italy

15:30-15:50 **Femtosecond time resolved spectroscopy of 9-fluorenone and cyclopropylidene,** Juliane Köhler, University of Wuerzburg, Germany

16:10-16:30 **Time resolved studies of catastrophic optical mirror damage in red/NIR laser diodes,** Stella N. Elliott, Cardiff University, Cardiff, United Kingdom

15:50-16:10 **Momentum-dependent snapshots of a melting charge density wave in TaS_2,** Stefan Kaiser, CFEL, Hamburg, Germany

16:30-16:50 **COFFEE BREAK**

16:50-17:10 **Coherent optical phonons in different phases of $Ge_2Sb_2Te_5$ upon strong laser excitation,** Javier Solís, Instituto de Óptica, CSIC, Madrid, Spain

17:10-17:30 **Femto to nanosecond laser-induced damage in hafnia coatings**, Laurent Gallais-During, Institut Fresnel, CNRS, Marseille, France

17:30-17:50 **Attosecond Stark Spectroscopy on molecular systems,** Franck Lépine, Université de Lyon, France

17:50-18:10 **Thin-film-metallic-photocathodes grown by pulsed laser ablation**, Antonella Lorusso, University of Salento, Lecce, Italy

20:00 **GALA DINNER**

TUESDAY, MARCH 29th

Session 3. Advanced Laser Technologies for the Progress of Chemical and Life Sciences

Chair: Oldrich Renner, PALS, Prague, Czech Republic

09:00-09:30	**Keynote presentation, Advanced inspection of matter using laser spectroscopy. From laboratory to extreme scenarios**, Javier Laserna, Universidad de Málaga, Málaga, Spain
09:30-09:50	**Characterization and Control of Nanostructures on Polymers and Biopolymers by Laser Irradiation**, Esther Rebollar, Instituto de Química Física Rocasolano, CSIC, Madrid, Spain
09:50-10:10	**Conical intersection dynamics in Rhodopsin and Isorhodopsin**, Philipp Kukura, Oxford University, Oxford, United Kingdom
10:10-10:30	**Binding of anthracyclines to a G-quadruplex structure of telomeric DNA and excited state deactivation processes in the complexes**, Ilse Manet, ISOF-CNR, Bologna, Italy
10:30-10:50	**COFFEE BREAK**
10:50-11:10	**Electronic Relaxation and Vibrational Wavepacket Motion in Poly(3-hexylthiophene) Films Studied by 2D Electronic Spectroscopy**, Arvydas Ruseckas, University of St Andrews, United Kingdom
11:10-11:30	**Proton coupled electron versus hydrogen atom transfer in BLUF photoreceptors**, Tilo Mathes, Humboldt University, Berlin, Germany
11:30-11:50	**Noninvasive observation of skeletal muscle contraction using near- infrared time-resolved reflectance and diffusing-wave spectroscopy**, Thomas Gisler, Universität Konstanz, Konstanz, Germany
11:50-13:30	**ROUND TABLE chaired by Annie Klisnick (Université Paris-Sud, Orsay, France) and Rosa Weigand (Universidad Complutense, Madrid, Spain)**
13:30	**CLOSING REMARKS, END OF MEETING**

LUNCH AND OPTIONAL VISITS TO INO LABS IN PISA AND INO AND LENS IN FLORENCE

Session 1. Interactions at Extreme Energy Density

Chair: Antonio Giulietti, INO, Pisa, Italy

The collaboration of European FEL facilities

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X-ray Free Electron Lasers (FELs) have made enormous progress in the last decade. FLASH at DESY has been operating as the world's first soft X-ray FEL user facility since 2005. LCLS in the USA started user operation in 2009 and delivers few-femtosecond hard X-ray pulses down to 1.5 Angstrom wavelength. New X-ray FEL sources in Europe and overseas are starting operation or are under development, such as Fermi@Elettra, SPring-8 XFEL in Japan, European XFEL, SwissFEL, SPARX and MAX IV FEL.
The European stakeholders realised early on that a close collaboration was needed in order to meet the technological and scientific challenges of these novel and rapidly developing technologies and to fully exploit the potential of these unique short-pulse X-ray sources. The first steps were made in the EUROFEL Design Study which was funded under FP6 and included all interested European stakeholders.
The importance of free electron lasers for research and innovation in Europe was also recognised on the political level, and two FEL projects were thus included in the ESFRI Roadmap 2006, the European XFEL as an international facility and IRUVX-FEL as a consortium of national facilities. Subsequently, the preparatory phase of the two ESFRI Roadmap projects, PRE-XFEL and IRUVX-PP, were supported by the European Commission and will end 30th June 2011 and 30th March 2011, respectively. While PRE-XFEL prepared the construction of the European XFEL in Hamburg, IRUVX-PP focused on defining and structuring a close long-term collaboration of all European FEL and accelerator based short-pulse facilities. The presentation will give an outline of these activities and the future perspectives.

Optical field ionized XUV laser using the guiding of the pump beam in a plasma performed channel

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We employed the guiding of pump beam by the preformed plasma channel for efficient pumping of optical field ionized XUV laser in a dense krypton jet. First the profile of electron density of the plasma channel in high density Kr gas jet, created by two consequent laser pulses, was measured interferometrically, in order to achieve the best conditions for guiding the pump beam and produce longer gain medium. Very strong lasing of the Ni-like krypton at the wavelength of 32.8nm was achieved using 1cm long gas jet with backing pressure of 100 bar due to the efficient propagation of the pump beam through the dense media (the estimated density was up to 10^{20} cm^{-3} which is two orders of magnitude higher than the optimum density of the gas cell when no guiding was employed [2]). As the gain duration of the collisionally excited XUV lasers strongly depends on collisional ionization that decreases the population of lasing ions, we suppose that increasing the plasma density the gain duration gets shorter and calculations predict the sub-100fs duration for densities around 10^{20} cm^{-3}. We used the variable path-difference interferometer to estimate the spectral profile of the produced beam and hence to determine the Fourier limit of the XUV pulse duration.

References

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Electron beams and X-ray radiation generated by laser wakefield in capillary tubes

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Intense short duration laser pulses interacting with light gas targets expel electrons from the regions of high intensity and leave in their wake a plasma wave. This wave is associated to space charge electric fields used to accelerate relativistic electrons in the so called laser wakefield mechanism. In the non linear regime of laser wakefield, plasma electrons can be completely blown out of the intense laser region, and self-trapped in the accelerating potential of the plasma wave. Accelerated electrons are also submitted to transverse fields which make them oscillate transversely during the acceleration process, and they emit radiation similar to synchrotron radiation.
Non linear laser wakefield and the set-up of the blown-out regime were studied in long plasma targets during experiments performed at the Lund Laser Centre (LLC). In order to extend the length of interaction of the intense laser with the plasma, capillary tubes with length 10 to 30 mm were used as waveguides. The laser propagation is governed, in the non linear regime, by the interplay between self-guiding and reflection at capillary walls. Using the LLC 10-Hz Ti-sapphire pulsed laser with 20 TW on target, electrons were accelerated to the 200 MeVs range, and bright X-ray beams with synchrotron spectra with peak energy in the range 2-4 keV were produced. A large range of parameters was explored to study the role of the capillary tube, and extend the plasma length to the limit of dephasing, when the electrons outrun the accelerating fields. Preliminary results will be presented and compared to 3D PIC simulations.

Measurement of harmonics produced in gas-puff laser plasmas by synthetic single-crystal diamond detectors

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We report on the experimental results obtained by the use of VUV diamond detectors for a measurement of harmonic radiation produced by interaction of 248-nm KrF laser radiation with gas-puff argon plasma. The experiment was performed at the University of Szeged in Hungary. Energy of laser pulses was about 20 μJ and duration of the pulses 600 fs.
Due to the high discrimination ratio of diamond based detectors between the fundamental KrF laser radiation and its harmonics, an accurate measurement of the produced harmonic content was possible.
Two different structures based on high quality Chemical Vapour Deposition (CVD) single crystal diamond produced in Rome "Tor Vergata" University laboratories were tested. The first one is a Schottky diode in a multilayered p-type diamond/ intrinsic diamond/ Schottky metal configuration with a semitransparent top electrode and operating in transverse configuration. The second one is a photoconductive detector based on a nominally identical single crystal CVD diamond film, having interdigitated electrodes and operating in planar configuration. Both devices were electrically characterized by I-V measurements and tested in the EUV spectral region by using He-Ne DC gas discharge radiation sources and a toroidal grating vacuum monochromator. An optical parametric oscillator tunable laser was also used to investigate the visible-blind properties of the photodetectors by measuring the photoresponse at different wavelengths in the 210–500 nm range.
In the preliminary experiment on harmonics in Szeged, very promising results were obtained. Using a diamond detector and sensitive charge preamplifier, harmonics up to the 7-th one were measured (behind a monochromator) and the third harmonics was registered directly from the gas-puff target at presence of the fundamental radiation.

Proton acceleration and X-ray emissionin fast electron transport studies

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Generation and transport of so-called fast electrons driven by ultraintense laser pulses are the key to inertial confinement fusion (ICF) in the fast ignition (FI) approach [1], the baseline approach of the HiPER project [2]. Fast electrons also provide the basic mechanism for emission of Ka ultrashort X-ray pulses [3]. Moreover, fast electrons play a crucial role [4] in laser driven ion acceleration with target normal sheath acceleration (TNSA) mechanism. In these interactions fast electron currents are well above the Alfven limit and therefore transport strongly depends upon the conductivity of the medium which must support propagation by supplying a balancing cold electron return current. Several mechanisms can influence propagation and affect transport of fast electrons across the target material. Among these mechanisms it was predicted that when a fast electron beam with some angular spread is normally incident on a resistivity gradient, as that available in layered targets, magnetic field generation would occur that could inhibit beam propagation [5]. There is little experimental evidence of this and measurements are often affected by uncertainties on the laser pulse features or target defects like vacuum gaps between layers.

We use well characterized laser pulses and lacquer dielectric coated [6] targets to rule out vacuum gaps between metal and dielectric and we measure proton emission from laser irradiated coated and uncoated metal foils. We find conclusive evidence[7] that such transport effects occur in this scenario. We also use novel [8] X-ray imaging techniques to study front and rear side X-ray emission from thin titanium foils under the same irradiation conditions. Significant differences in intensity, dimension, and spectrum between front and rear side emission intensity in the 3-12 keV photon energy range was found even for 5 μm thin Ti foils.Simulations and analysis of space resolved spectra show that this behavior can be interpreted [9] in terms of directional Bremsstrahlung emission from fast electrons generated during the interaction process.

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Radiative Shocks at PALS

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Since 10 years, the study of strong radiative shocks waves in gases has been stimulated by the availability of kJ laser installations, which allow to launch shock waves with velocities of about 60 km/s [1-3] or more [4] in gases, and then to study the coupling between radiation and hydrodynamics in simple geometries. The topic is important for the understanding of the physics and radiative signatures of astrophysical shock waves, like those occurring during the stellar formation.
Radiative shocks are characterized by an ionization front induced by the shock wave. For a given shock velocity, which is set by the available laser energy, and a given initial gas pressure, the radiative effects are more important for gases with high atomic number. A laser fluence of 10^{14} W/cm^2 on a thin foil is sufficient to drive a shock wave at an initial velocity of about 60 km/s in Xenon at 0.1-0.3 bars. I will present recent results obtained on the PALS laser facility in Prague, in the frame of a collaboration between Observatoire de Paris, Laboratoire de Physique des Plasmas, CEA, Imperial College and Institute of Physics/PALS Center.
The financial supports from Laserlab Europe, CNRS PICS4343 and ANR-08- BLAN- 0263-01 are acknowledged.

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~2.1 µm laser based on Ho:KLu$(WO_4)_2$ crystal in-band pumped by a Tm:KLu$(WO_4)_2$ laser and a diode laser

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In the last years, infrared solid-state lasers based on Ho^{3+} ions (Ho) operating slightly above 2 µm have been the subject of intensive research, either by co-doping the active medium with Tm^{3+} ions (Tm) or by direct excitation using resonant or in-band pumping of the $^5I_7$ excited level of Ho at ~1.9 µm. The latter offers the advantages of high quantum efficiency for the ~2.1 µm laser transition and minimal heat load to the active element, both important prerequisites for power scaling in this spectral range.

The monoclinic potassium double tungstates with the general formula KRE$(WO_4)_2$ (KREW) where RE is a passive trivalent ion (Y, Gd, or Lu), stand out because of their very high transition cross sections (absorption and emission) and weak concentration quenching of the fluorescence which is related to the relatively large dopant-to-dopant separation. The less known KLuW was recently shown to be especially suited for Yb and Tm doping.

In this work we demonstrate continuous-wave (CW) lasing of the Ho^{3+}-ion in a singly-doped KLuW crystal at room temperature and under in-band pumping using two different pump sources, a diode-pumped Tm^{3+}:KLuW laser and a diode laser.

With diode-pumped Tm^{3+}:KLuW laser pumping, the maximum achieved slope efficiency of the Ho:KLuW laser amounts to ~55% with respect to the absorbed power and the maximum output power of 648 mW is generated at 2078 nm. These results reveal promising potential of this new laser material [1]. No thermal roll-off in the power dependence and no damage to the uncoated active element have been observed for the available pump power.

With a fiber coupled GaSb diode stack as pump source operating at ~1941 nm, a maximum output power of 408 mW and a slope efficiency of 54.5% with respect to the absorbed pump power were achieved at ~2080 nm for Ho:KLuW. A comparison of the laser performance within the Ho:KREW family of crystals showed a slightly superior performance of Ho:KYW, achieving a maximum output power of 438 mW and a slope efficiency of 58.8% [2].

In conclusion, we demonstrated CW laser operation in resonantly pumped Ho:KREW crystals with two different pump sources showing promising laser performance for power scaling. Such scaling along with passive *Q*-switching by a saturable absorber are experiments planned for the next future.

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Three-dimensional photonic quasicrystals

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Three-dimensional photonic quasicrystals not only appeal due to their geometrical beauty but also serve to study optical transport phenomena for materials with optical properties situated in the interesting region between ordered and disordered materials. So far, icosahedral quasicrystals are the only three-dimensional quasicrystal structures that have actually been realized for microwave [1] and optical frequencies [2]. Here, we present a rationally construction scheme based on the cut-and-project- method to devise rhombicuboctahedral three-dimensional quasicrystals. This is done by rotating a fictitious twelve-dimensional simple-cubic crystal and subsequently projecting it into the three-dimensional real space. In this way we calculate the lattice points for our structures. Coresponding polymer microstructures are fabricated by means of three-dimensional direct laser writing. The samples are characterized by scanning electron microscopy and Laue diffraction. The Laue diagrams taken with visible light reveal the theoretically anticipated unusual eight-fold rotational symmetry [3].

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Session 2. Ultrafast Laser Probing of Matter

Chair: Marta Castillejo, CSIC, Madrid, Spain

Probing primary photoinduced processes in organic molecules with tunable few-optical-cycle light pulses

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Many light-induced processes in organic molecules, such as energy relaxation, energy/charge transfer and conformational changes, occur on ultrafast timescales, ranging from 10^{-14} to 10^{-13} s. The speed of such elementary processes is intimately linked to their efficiency, making ultrafast optical spectroscopy an invaluable tool for their investigation, since it allows tracking in real time the energy flow following photoexcitation. Pump-probe spectroscopy requires both short pulses, in order to observe fast dynamics, and broad frequency tunability, to excite a system on resonance and probe optical transitions occurring at different frequencies. Optical parametric amplifiers (OPAs) are ideal tools for such experiments, because they provide frequency tunability and support broad gain bandwidths, enabling the generation of very short pulses [1].
In this talk we will describe a state of the art system, based on two synchronized OPAs, providing sub-10-fs temporal resolution over a very broad spectral range, from 400 nm to 2 μm. After reviewing the pulse generation techniques and the system performance, we will present selected examples of applications to the study of ultrafast processes, such as: energy transfer in photosynthetic light harvesting complexes, electronic and vibrational dynamics in carbon nanotubes, isomerization of rhodopsin [2].

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Femtosecond time-resolved spectroscopy of 9-fluorenone and cyclopropylidene

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This study of the photophysics of fluorenone is motivated by the central role of this chromophore in two classes of molecules. The first class are polyfluorenes, considered for various optoelectronic applications. Here the oxidative degeneration of single fluorenes to fluorenone changes the emission properties of the polymer. The second class are truxenones, which are of interest as materials for nonlinear optics.

We recently investigated the ultrafast dynamics of isolated fluorenone by femtosecond time-resolved photoionization and photoelectron spectroscopy. Excitation at 266 nm is followed by a two-step internal conversion to the long-lived S_1 state. A corresponding shift of the photoelectron spectrum to lower kinetic energies is observed.

In addition we studied the femtosecond dynamics of the $^1B_1$ state of cyclopropenylidene (c-C_3H_2), produced by flash pyrolysis. The reactive singlet carbene was previously investigated in our group utilizing REMPI and Doppler spectroscopy. Broad bands in the UV spectrum imply an ultra-short lifetime, which motivated us to examine the reactive intermediate by time-resolved photoelectron spectroscopy.

Time resolved studies of catastrophic optical mirror damage in red/NIR laser diodes

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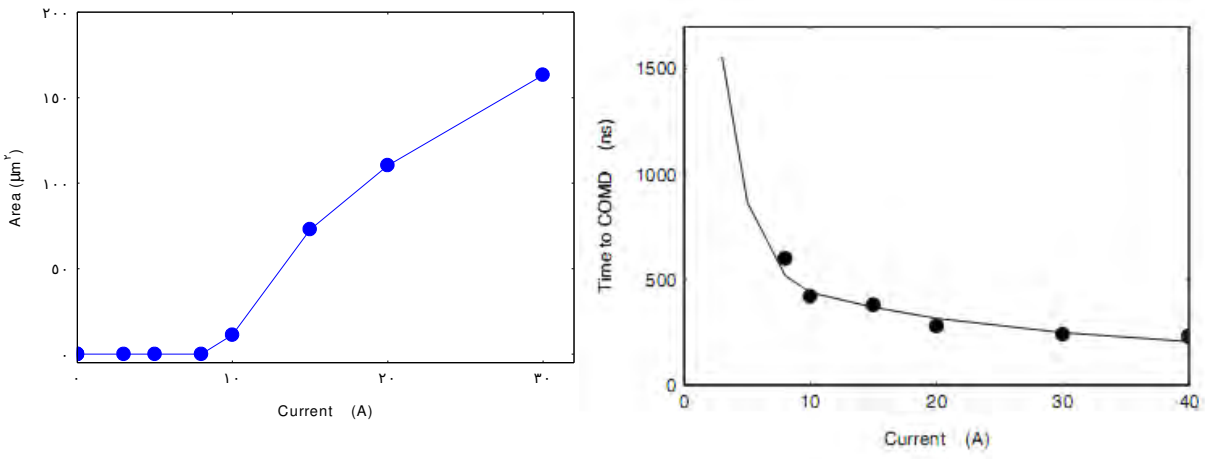
Catastrophic optical mirror damage (COMD) remains a limiting factor in the increasing drive for high power semi-conductor diode lasers. The total COMD process consists of an initial facet temperature rise, followed by thermal runaway, facet damage and propagation of a damaged or molten region leading to dark lines in electroluminescence viewed in the plane of the active region, with an abrupt drop of laser power.

We have observed for the first time in real time the changing light intensity during catastrophic optical mirror damage in red emitting AlGaInP quantum well (QW) based and near infra-red emitting quantum dot (QD) laser diodes. Using as-cleaved facets and this material system, which is susceptible to COMD, we applied single, high current pulses of up to 40 A, recording the drop in light intensity on the timescale of tens of nanoseconds. Microscope images of the facet before and after the current pulse confirmed severe damage, with an area of damage to the facet that showed a clear increase with drive current. (figure 1(a)). We found for QW the total COMD process up to the drop of light intensity to non-lasing levels (t_{COMD}) takes place on a timescale of hundreds of nanoseconds, approaching a limiting value of 200 ns (figure 1(b)). We also carried out scanning electron microscopy and cathodoluminescence (CL) on selected devices. We found the behaviour of QD devices to be quite different, with little facet damage evident, and a measured t_{COMD} that approached zero asymptotically.

Using a straightforward energy model[1] we explained the limiting time at high currents and the relationship between the time to COMD and the area of damaged facet material for QW devices. The total time to COMD (t_{COMD}) is the sum of the time for the temperature to rise (t_{rise}) and the time to melt (t_{melt}). t_{rise} is dominated by the relatively slow facet temperature rise to the critical temperature at which thermal runaway occurs, the time for runaway being negligible in comparison. t_{rise} and t_{melt} depend on the optical power and the mass of heated or damaged material. Assuming temperature rise and melting rates proportional to the optical power, and therefore approximately inversely proportional to current, we obtained a fit to the measured t_{COMD}.



(a) (b)

Figure 1. (a) Measured area of damage on facet of QW devices as a function of current. (b) Measured t_{COMD} (points) and fit from the energy model (line).

We then considered a possible physical basis for the values of the fitting parameters. We obtained CL images of the active region after damage which gave a value of 5–13 μm/μs for the propagation velocity of the damaged region inside the cavity enabling the volume of damaged material inside the active region to be estimated. With some assumptions we then obtained reasonable estimates for the fitting parameters in terms of quantities of material involved and latent and specific heats, thus supporting our energy argument.

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Momentum-dependent snapshots of a melting charge density wave in TaS_2

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Charge density waves (CDWs) underpin the electronic properties of many complex materials. Near-equilibrium CDW order is linearly coupled to a periodic, atomic-structural distortion, and the dynamics is understood in terms of amplitude and phase modes [1]. Using photoexcitation in a 2D Mott and CDW system 1T-TaS2 we map out the ultrafast response of electronic correlations and order in the Mott and CDW state using time and angle resolved photoemission (ARPES) with sub 30 fs XUV pulses. Besides an ultrafast melting of the Mott gap, known from 6 eV ARPES experiments [2] and which we can map out in the whole Brioullin zone now, we also find an ultrafast response of the electronic order in the CDW, which melts before the underlying structural distortion relaxes. This decoupling of charge and lattice modulation challenges the view of Fermi surface nesting for the CDW formation and the hierarchy of electronic order in this system.
The experiments reported here employed the time-resolved laser ARPES setup at the Artemis facility, in the Central Laser Facility of the Rutherford Appleton Laboratory. A 1-kHz pulsed beam of 30-fs pulses from an amplified 800-nm laser is split into two parts. One is used as the pump, that is, to drive the system out of its equilibrium state. The other enters a beamline held under vacuum, and is focussed into a pulsed jet of argon gas in order to generate XUV pulses via the non-linear optical process known as high-order harmonic generation. The XUV passes through a grating monochromator designed to preserve the pulse duration [3], which selects the 13^{th} harmonic (photon energy 20.4 eV). A grazing-incidence toroidal mirror focuses the pulsed XUV onto the sample, which is held on a liquid-helium-cooled sample manipulator in a UHV chamber. Photoelectrons emitted from the sample surface after each XUV pulse are collected and measured by a hemispherical analyser (SPECS PHOIBOS 100) with a multi-channel plate and CCD detector. The overall energy resolution of the system is approximately 150 meV, limited by the line width of the laser XUV source.

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Coherent optical phonons in different phases of $Ge_2Sb_2Te_5$ upon strong laser excitation

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The response of $Ge_2Sb_2Te_5$ films to intense femtosecond laser pulses is studied by optical coherent phonon spectroscopy. The three different phases of this phase-change material (amorphous, fcc- and hcp-crystalline) are investigated, featuring different carrier and coherent optical phonon (COP) dynamics. In addition, the COP dynamics of the amorphous film after laser crystallization has been measured. Satisfactory fitting of the reflectivity data can only be obtained if at least two main phonon frequencies are considered in the fit. The evolution of these frequencies for increasing pump energies is investigated for the fcc and the laser-crystallized phase. The results reveal a systematic red-shift of both frequencies in the fcc- phase, whereas they remain constant in the laser-crystallized film. A considerable fraction of amorphous phase remains in the laser-crystalline material, significantly distorting the phonon spectrum. These results are important for emerging strategies aimed at driving ultrafast phase transitions via coherent phonon excitation for applications in data storage.

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Femto to nanosecond laser-induced damage in hafnia coatings

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The damage of dielectric material by femtosecond laser has been studied in detail by the scientific community, thus providing the basis of mechanisms involved in this interaction. At the femtosecond scale only electronic processes can be excited by the electric field and contribute to the absorption of energy from the laser pulse. Moreover, this absorption occurs at a time scale very short compared to heat transfer time. It is possible to uncouple and treat separately the mechanism of absorption, energy transfer to the structure, and physical degradation of the material.
Despite common characteristics with bulk materials, the laser damage of dielectric thin films has peculiarities that need to be studied. Dielectric thin films have specific optical, mechanical, thermal and electronic properties, affecting the resistance of components under laser exposition. Indeed, a strong dependence of damage threshold with the deposition parameters used was observed in nanosecond regime. An investigation into femtosecond regime is therefore necessary to update the influence of various physical parameters such as deposition technique, the nature of the material used or the thickness.
In this work we report laser-damage thresholds and morphologies of hafnia single layers exposed under femtosecond, picosecond, and nanosecond single pulses (1030 and 1064 nm). The samples were made with different deposition parameters in order to study how the damage behavior of the samples evolves with the pulse duration and how it is linked to the deposition process. In the femtosecond to picosecond regime, the scaling law of the laser-induced damage threshold as a function of pulse duration is in good agreement with the models of photo and avalanche ionization based on the rate equation for free electron generation. However, differences in the damage morphologies between samples are shown. No correlation between the nanosecond and femtosecond/picosecond laser-damage resistance of hafnia coatings could be established. We also report evidence of the transition in damage mechanisms for hafnia, from an ablation process linked to intrinsic properties of the material to a defect-induced process, that exists between a few picoseconds and a few tens of picoseconds.

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Attosecond Stark Spectroscopy on Molecular Systems

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Stark spectroscopy is a well-established technique that is used to derive information on molecular structure and chemical bonds. In this approach, a molecular sample experiences a macroscopic static (or oscillatory) electric field created by electrodes, which polarize the molecules and therefore induce a Stark shift that modifies their electronic states. The variation of resonant light absorption provoked by the Stark effect is measured and direct information on the nature of the molecule is extracted from this variation. With short laser pulses the strength of the instantaneous electric field of the light can be considerably higher than what is usually produced at a macroscopic scale. In addition, the ultrafast oscillation of the field makes it possible to reach a situation where not only the DC Stark effect but also electron dynamics in the field play a role. As a consequence, using light instead of macroscopic electrodes may offer the possibility to extend the Stark spectroscopy technique, provided that the ultrafast variation of the light electric field can be observed. That is exactly what attosecond science has recently achieved. Therefore, modern attosecond beamlines offer the adequate tools to develop attosecond Stark spectroscopy.
We have performed experiments using the attosecond pump-probe set-up developed at the LLC. We have studied the influence of the instantaneous electric field of the IR fs pulse on various molecules (for simple diatomic N_2, O_2 and large $C_{14}H_{10}$ PAH), where the light-induced Stark effect is "read" by XUV attosecond pulses. We will discuss insights that can be obtained on electronic states, the influence of nuclear motion and electron dynamics at the attosecond timescale. We will discuss applicability and further developments of this new technique.

Thin-film-metallic-photocathodes grown by pulsed laser ablation

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In this paper we review the current status of metallic photocathodes based on thin films prepared by pulsed laser deposition (PLD), a very efficient technique for producing adherent and uniform thin films [1], exploring ways to improve the performance of these devices. Photocathodes based on Mg and Y thin films have been characterized by different diagnostics techniques and tested in a photodiode cell to deduce their photoelectron properties. The quantum efficiency (QE) of our photocathodes was systematically improved by laser cleaning treatments reaching the corresponding bulk values [2]. The level and the vacuum quality have an unquestionable influence on both, the QE value and lifetime, achievable on metallic-film-based cathodes. Time-resolved-mass-spectrometric investigations definitively suggest that the deposition of high purity metallic thin films should be carried out after a deep and careful laser cleaning of the target surface and in ultra-high-vacuum systems. This laser cleaning is highly recommended not only to remove the first contaminated layers but also to improve the quality of the vacuum, by reducing the partial pressure of contaminant chemical species as H_2 and O_2 molecules. The rule of the metallic photocathode adsorbed gases on the emission performance will be also reported and discussed.

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Session 3. Advanced Laser Technologies for the Progress of Chemical and Life Sciences

Chair: Oldrich Renner, PALS, Prague, Czech Republic

Advanced inspection of matter using laser spectroscopy. From laboratory to extreme scenarios

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The Laser Laboratory of University of Malaga is a research infrastructure funded by regional, national and international programs in chemistry and materials research. The facility is specialized in the development and application of advanced laser technologies for materials characterization and offers quick-turn-around, cost-effective solutions to a variety of research and technical problems in diverse applications areas, including microelectronics, process monitoring technologies, environment, cultural heritage, security, and steel products and processes. Competencies span from inspection of nanodomains to teledetection using a variety of laser systems. Experiments at the Laser Laboratory include fs laser-ionization time-of-flight mass spectrometry with simultaneous observation and spectral analysis of desorbing plume. A new system is being installed for fs-time resolved microscopy for inspection of ultrafast laser ablation processes. The facility also features secondary ionization mass spectrometry for isotope ratio measurements and for surface molecular and atomic imaging. The Laboratory also designs and builds complete spectroanalytical systems to custom specifications, including compositional mapping systems, remote analysis instruments and portable/field deployable prototypes. Systems for underwater laser analysis of materials and for standoff inspection of materials at distances up to 200 m are in operation. In this talk, an overview of the Laboratory activities will be presented. The most significant results will be also discussed.

Characterization and control of nanostructures on polymers and biopolymers by laser irradiation

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Processing techniques based on laser irradiation and ablation provide high precision at the micro- and nanoscale, are suitable for all types of materials and afford accuracy and control to fabricate new nanomaterials with applications in photonics, sensing, biomedicine, and others [1]. The use of ultrashort femtosecond (fs) laser pulses is highly attractive for the micro- and nanostructuring, mainly because the pulse durations of these lasers are shorter than typical material relaxation times, and therefore the laser–material interaction, rather than the material thermal properties, generally determines the outcome of the laser fabrication process. Fabrication of micro- and nanostructures on polymers and biopolymers, is of high interest in technological areas of organic photonics, biomedicine and bioengineering [2]. Biopolymers hold promise to become the fundamental building blocks for organic photonics and electronics; in biomedicine, laser processing of these materials allows the fabrication of biocompatible porous scaffolds for cell growth and drug release. Ultrashort laser pulses of fs duration improve the spatial resolution and reduce the mechanical and thermal deformation as compared with longer pulses.
In this work, self standing films of chitosan, starch and their blend were irradiated in air with single UV laser pulses of different duration using a KrF excimer (248 nm, 20 ns), a Nd:YAG (5th harmonic, 213 nm, 150 ps) and a hybrid excimer-dye (248 nm, 500 fs) systems. On line control of the irradiated areas was performed by monitoring the fluorescence induced at low fluence with the same irradiation laser, while the modifications of surface morphology were observed by scanning electron microscopy. Nanostructuring of the surface in the form of pores and bubbles is observed in all samples irradiated with 500 fs pulses. However when longer ps and ns pulses are applied, this type of superficial morphology only develops in the case of chitosan. These differences are related to the mechanisms operating under the different pulse temporal domains and to the material properties, particularly absorption coefficient and thermal parameters which determine the amplitude of the laser-induced pressure wave.

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Conical intersection dynamics in Rhodopsin and Isorhodopsin

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The primary visual event, the cis-trans isomerisation of the the retinal chromophore in the visual pigment rhodopsin is one of the most rapid and efficient photochemical reactions in nature. Its unique reactive properties have been proposed to be mediated by a conical intersection connecting electronic excited and ground state thereby allowing for fast and directed decay of the initial light-induced excitation towards the photochemical product. We have used ultrashort pulses ranging from the visible to the near-infrared to probe the reactive dynamics with unprecedented energy and temporal (<20 fs) resolution. Our results are clearly indicative of a system passing through a conical intersection and in combination with theoretical modelling, provide a real-time molecular movie of this prototypical photochemical reaction. The results in rhodopsin are compared with those obtained for Isorhodopsin, the 9-cis retinal analogue of the 11-cis retinal chromophore found in rhodopsin, exhibiting similar, but much slower and less efficient passage through a conical intersection.

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Binding of anthracyclines to a G-quadruplex structure of telomeric DNA and excited state deactivation processes in the complexes

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Anthracycline drugs are thought to exert their action via the formation of a complex with duplex DNA through intercalation. Little is known about their interaction with guanine-rich sequences in their G-quadruplex form. G-Quadruplex structures have drawn a lot of attention during the last decade due to the emerging evidence that they are involved in several biological functions comprising also tumor development and progression. Considering the interest in G-quadruplex structures as potential target in cancer therapy and the affinity of the drug for GC base pairs we started an investigation of the drug affinity for G-quadruplex structures. Telomeres are made of guanine-rich sequences believed to organize in G-quadruplex structures and some literature data report that anthracycline drugs are able to interfere with the DNA telomere maintenance processes important for cell survival. We thus decided to study the interaction of two anthracyclines with telomeric DNA G-quadruplex structures. In particular we have observed the formation of complexes of 1:1 and 2:1 stoichiometry of doxorubicin and MEN10755, a doxorubicin derivative with two sugar units, with G-quadruplex structures of the telomeric DNA sequence, 5'-d[AGGG(TTAGGG)$_3$]-3' in different media. The fluorescence of the drug in the complex is quenched in K^+ rich solution while the 2:1 complex maintains some fluorescence in Na^+ rich solution. In order to clarify the mechanism underlying the fluorescence quenching we investigated the photophysical properties of these complexes on fs-ps time-scale. From the thermodynamic point of view photoinduced electron transfer is exergonic for both drug and nucleotide in the excited singlet state. We will illustrate the results obtained up to now.

Electronic Relaxation and Vibrational Wavepacket Motion in Poly (3-hexylthiophene) Films Studied by 2D Electronic Spectroscopy

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Conjugated polymers show great potential for development of solution-processable low cost and large area solar cells. Photovoltaic cells made of regioregular poly(3-hexylthiophene) (P3HT) blended with fullerene derivatives show nearly 100% internal quantum efficiency but the mechanism of efficient charge separation is not understood yet. We have studied ultrafast excitation dynamics in P3HT films using two-dimensional electronic spectroscopy. With site-selective excitation at the low energy tail of the absorption spectrum we observe spectral dynamics on a time scale from 20 to 300 femtoseconds which can be attributed to intra-band exciton relaxation in a disordered H-aggregate formed by strong electronic coupling between conjugated chains with π - π stacking. Broadband excitation using 15 fs pulses shows oscillations of the 2D signal amplitude which survive up to a picosecond and indicate a persistent vibrational coherence. We discuss the impact of primary excitation dynamics on charge separation in photovoltaic structures.

Proton coupled electron versus hydrogen atom transfer in BLUF photoreceptors

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BLUF domains belong to the family of flavin containing blue light photosensory receptors[1]. Among these they show the least structural transitions in forming the biological active signaling state. In less than a nanosecond after excitation merely a hydrogen bond switch occurs between the flavin and its surrounding amino acids after sequential electron and proton transfer from a nearby tyrosine and subsequent radical recombination[2]. The switch is either accomplished by the reorientation and/or tautomerisation of a conserved glutamine side chain and indicated by a red shift of the S_0-S_1 absorption and a downshift of carbonyl vibrations of the flavin. The switch is reverted within a few seconds in the dark. We investigated the differences between light and dark adapted states by ultrafast spectroscopy on a mutated Slr1694 protein. The W91F mutation does not affect the ultrafast dark state photodynamics but increases the lifetime of the light adapted state ~10fold[3]. We used this behaviour to accumulate the light adapted state conveniently by background illumination and performed ultrafast visible and infrared absorption spectroscopy. In contrast to the dark state, where we observe a highly heterogenous excited state decay, the complexity is strongly reduced here. Moreover, the decay as opposed to the dark reaction occurs in a highly H/D isotope dependent manner. The primary product after excitation consists of a neutral flavin semiquinone according to vis spectroscopy. By ultrafast mid IR spectroscopy we firmly assigned the intermediate as a neutral radical pair consisting of a tyrosyl radical and a flavin semiquinone, which is similar to the neutral intermediate in the dark state reaction. The semiquinone intermediate simply decays back to the light state without any photodeactivation. Apparently the reactivity in the light adapted state is modified in favor of hydrogen atom transfer in contrast to sequential electron and proton transfer in the dark state.

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Noninvasive observation of skeletal muscle contraction using near-infrared time-resolved reflectance and diffusing-wave spectroscopy

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We introduce a novel method for noninvasively measuring muscle contraction *in vivo*, based on a combination of near-infrared time-resolved spectroscopy (TRS) and diffusing-wave spectroscopy (DWS). DWS exploits the information about time-dependent shear motions within the contracting muscle that are contained in the temporal autocorrelation function $g(1)(t,\tau)$ of the multiply scattered light field measured as a function of lag time, τ and time after stimulus, t. The analysis of $g(1)(t,\tau)$ measured on the human *M. biceps brachii* during repetitive electrical stimulation, using optical properties measured with TRS, shows that the tissue dynamics giving rise to the speckle fluctuations can be described by a combination of diffusion and shearing. The evolution of the tissue Cauchy strain e(t) shows a strong correlation with the force, indicating that a significant part of the shear observed with DWS is due to muscle contraction. The evolution of the DWS decay time shows quantitative differences between the *M. biceps brachii* and the *M. gastrocnemius*, suggesting that DWS allows to discriminate contraction of fast- and slow-twitch muscle fibers.

Posters

Influence of Laser-Plasma Parametric Instabilities in Shock Ignition Relevant Regime

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Abstract.
In the context of the Inertial Confinement Fusion Shock Ignition an experimental campaign at Prague Asterix Laser System (PALS) has been performed to study shock generation and laser-plasma coupling at intensity of the order of 10^{16}W/cm^2. During the experiment two beams have been used, the first one to create the extended preformed plasma (scale length of the order of hundreds um), the second pulse was used to create the shock properly delayed with respect to the first beam. The experiment was mainly focused on the studies of the shock breakout generated from a planar target, on laser-plasma coupling and parametric instability development. This document is focused on the data, and their analysis, collected with a optical back-scattering diagnostic devoted to measure the reflected energy, and to characterize the parametric instabilities as Brillouin, and Raman. Calorimetry and spectroscopic measurements allowed the study of the laser-plasma coupling in relevant Shock Ignition intensity regime. Based on spectral measurements of back reflected light an almost well defined region for laser energy absorption has been identified. Moreover, experimental data have shown that parametric instabilities do not play a strong role on the laser plasma coupling. By monitoring the back reflected light from the interaction region we found that only less than 5% of the total incoming laser energy was back reflected, and that only a little part of that light was coming from parametric instabilities.

The Laserlab ACCESS program at PHELIX

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PHELIX[1] is a high-energy, dual front-end laser system in operation at the GSI Helmholtz center for heavy-ion research in Darmstadt, Germany. In its long-pulse configuration, users can exploit frequency-doubled nanosecond pulses at an energy above 150 J in the UNILAC experiment hall, where ions with an energy up to 13 MeV/u are also available. Alternatively, the laser in its sub-picosecond pulse configuration is used for stand-alone experiments at 0.3 PW in the PHELIX hall (150 J, 500 fs). Two other target chambers are used for experiments with short pulses: one chamber is connected to the pre-amplifier output for experiments at moderate energy levels; while recently, a new compressor has been set up in the UNILAC experiment hall that can accept up to 100 TW laser pulses.
A new feature of PHELIX has been the possibility of working with two beams, made possible through the use of the 30-cm clear aperture of the main amplifier. In such a case, two beams with independent control over their relative delay and energy are simultaneously amplified in the amplifier chain. In a recent experiment, two 100 TW pulses were independently sent through the main amplifier and focused using two parabolas.
PHELIX acts as a free-access international facility where users get beam time based on the recommendation of an annual proposal-review committee (PPAC), which inspects access requests for the GSI facilities in the field of plasma physics. The PPAC also acts as the Laserlab experiment review committee. All proposals are evaluated in view of their potential scientific outcome. In addition, the PPAC favors experiments that either make use of both PHELIX and the ion facilities of GSI or contribute to the scientific program of PHELIX.
The scientific program of PHELIX is coordinated by the plasma physics department of GSI. In addition, it gets support from several German institutions in the framework of the upcoming facility for anti-proton research – FAIR[2]. These institutions are the Helmholtz institute Jena, the extreme matter institute (EMMI) and the Helmholtz international center for FAIR. The current topics of interest range from the injection of laser-accelerated ions into conventional accelerator structures, x-ray source development and the stopping power of laser-generated plasma. External users interested in such topics are invited to work in close collaboration with GSI for their proposal submission to the PPAC.

References

[1] V. Bagnoud et al., Appl. Phys. B **100** (1) 137-150 (2010)
[2] http://www.fair-center.org

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