Laserlab-Europe

User Meeting

29-30 November 2018

Paris, France

Book of Abstracts

Laserlab Europe User Meeting 2018

28 November 2018	
14:30-17:00	Optional tour of the APOLLON and ATTOLAB laser facilities (from/to Massy-Palaiseau RER station)
29 November – Charpak amphitheater, Jussieu campus, Sorbonne university	
09:30-09:40	Welcome addresses
09:40-10:10	The Laserlab-Europe TransNational Access Activity Didier Normand, Chair of the Laserlab-Europe Access Board
	Session I: *Imaging techniques & Technologies I* Chair: Rosa Weigand, Universidad Complutense, Spain
10:10-10:30	Karel Zidek, TOPTEC, IPP, Czech Republic "*Compressive Imaging of Transient Absorption Dynamics on the Femtosecond Timescale*" (project performed at LLC)
10:30-10:50	Yannick Deshayes, Laboratoire de l'Intégration du Matériau au Système, France "*Packaging-induced stress analysis in high-power DFB laser diode at 975nm*" (project performed at MBI)
10:50-11:20	*Coffee Break*
11:20-11:40	Xavier Ropagnol, INRS-EMT, Canada "*Sub-terahertz radiation from large aperture photoconductive antennas excited from UV laser beam*" (project performed at DP-USZ)
11:40-12:00	Frank Rosmej, LULI, France "*The transition from discrete to continuous spectra in dense media: two channel high-resolution X-ray imaging of Rydberg-transitions*" (project performed at PALS)
12:00-13:00	*Round table*
13:00-14:20	*Lunch*
	Session II: *Life-oriented research* Chair: Ingo Fischer, University of Würzburg, Germany
14:20-14:40	Carlo Altucci, Università "Federico II" of Napoli, Italy "*Time-resolved study of the laser mediated interactions between specially designed model system peptides*" (project performed at LIDYL/SLIC)
14:40-15:00	Daniela Täuber, Leibniz Institute of Photonic Technology, Germany "*Imaging nanoscale aggregation of proteins ex vivo using the contrast in Förster resonance energy transfer obtained from 2D polarization fluorescence imaging (2D POLIM)*" (project performed at LLC)
15:00-15:20	Kris Vissenberg, University of Antwerp, Belgium "*Regulation of root hair growth in the model plant Arabidopsis thaliana*" (project performed at CUSBO)
15:20-15:40	Tim Schäfer, University of Göttingen, Germany "*Photoelectron circular dichroism of fenchone from multiphoton ionization with nanosecond laser pulses*" (project performed at ULF-FORTH)
15:40-16:20	*Coffee Break*
	Session III: *Condensed matter* Chair: Helder Crespo, University of Porto, Portugal
16:20-16:40	Manuel de Anda Villa, INSP, France *"Ultrafast structural dynamics probed by photoelectron spectroscopy"* (project performed at CELIA)
16:40-17:00	Dmitry Spasskiy, Skobeltsyn Institute of Nuclear Physics, Lomonosov Moscow State University, Russia "*Non-linear effect of the interaction of the self-trapped excitons under dense laser excitation in molybdates*" (project performed at CELIA)
17:00-17:20	Davide Bossini, Technical University of Dortmund, Germany "*Ultrafast manipulations of high-energy spin excitations*" (project performed at CUSBO)

Status: 18 November 2018

17:20-17:40	Vitali Nagirnyi, Institute of Physics, University of Tartu, Estonia "*Cooperative luminescence of Yb pairs in Li6Y(BO3)3 single crystals*" (project performed at VULRC)
16:20-17:40	Laserlab Access Board Meeting (LULI meeting room)
19:00	*Meeting Dinner*
30 November – Charpak amphitheater, Jussieu campus, Sorbonne university	
	Session IV: *Atomic - Molecular Physics & Chemistry* Chair: Oldrich Renner, Institute of Physics, Czech Republic
9:30-09:50	Patrick Dupré, Laboratoire de Physico-Chimie de l'Atmosphère, France "*Sensitive Sub-Doppler Metrology in Cavity: from C2H2 to HD*" (project performed at LLAMS)
09:50-10:10	Arne Senftleben, University of Kassel, Germany "*Photoelectron circular dichroism and laser-induced electron diffraction of chiral fenchone molecules*" (project performed at ICFO)
10:10-10:30	Devendra Mani, University of Bochum, Germany "*Dissociation of HCl at 0.4 K: Sequence Matters*" (project performed at FELIX)
10:30-10:50	Caterina Vozzi, CNR-IFN, Italy "*Time resolved photoelectron circular dichroism*" (project performed at FERMI)
10:50-11:20	*Coffee Break*
	Session V: *Imaging techniques & Technologies II* Chair: Jouko Korppi-Tommola, University Jyväskylä, Finland
11:20-11:40	Aleksei Murzanev, IAP RAS, Russia "*Measurements of fluence profile and plasma density in femtosecond laser filaments and superfilament in ambient air under different convergence of terawatt laser pulses*" (project performed at HIJ-FSU)
11:40-12:00	Milutin Kovacev, Leibniz University Hannover, Germany "*Nanoscale lensless stereo imaging using dual coherent soft X-ray beams*" (project performed at LIDYL/SLIC)
12:00-12:20	Mabel Ruiz-Lopez, DESY, Germany "*Schwarzschild objective alignment by means of wavefront diagnosis*" (project performed at LOA)
12:20-12:40	Michael Zuerch, HIJ, Germany "*Single Shot XUV Nanoimaging Using an Intense Femtosecond Soft X-ray Laser*" (project performed at LOA)
12:40-14:00	*Lunch*
	Session VI: *Plasma physics* Chair: Valdas Sirutkaitis, VULRC, Lithuania
14:00-14:20	Ross Gray, University of Strathclyde, UK "*Intense laser-energy coupling and partition in the transition to relativistically transparent laser-plasmas*" (project performed at GSI)
14:20-14:40	Francesco Barbato, University of Zurich, Switzerland "*Quantitative X-ray Phase Contrast Imaging of a laser driven shock wave*" (project performed at GSI)
14:40-15:00	Luca Volpe, Centro de Láseres Pulsados (CLPU), Spain "*Enhanced relativistic-electron beam collimation using two consecutive laser pulses*" (project performed at LULI)
15:00-15:20	Closing remarks

Compressive Imaging of Transient Absorption Dynamics on the Femtosecond Timescale

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Ultrafast spectroscopy provides us with essential information needed to understand carrier dynamics in many systems of interest, such as solar cells or light-harvesting complexes. The ability to carry out imaging of the measured dynamics shifts the potential even further. One can, for instance, imagine the possibility to map and identify the "leaking" points of a solar cell, where the carriers rapidly recombine. However, an implementation of the imaging in ultrafast spectroscopy is a complex issue, because it requires recording a piece of 3D information (2D image, 1D time) by using a single-pixel detector, such as a photodiode or a spectrograph. One option lies in carrying point-by-point scanning of a sample by using a standard setup, which can be prohibitively long in many cases. Another option is to rebuild the pump-probe setup and sacrifice a part of information, e.g. the spectral resolution.

In collaboration with Lund Laser Center, Sweden (laser laboratory of D. Zigmantas at the Dept. of Chem. Phys., Lund University), we built an experimental setup, which can record the full information about a sample (i.e. spectral, spatial and temporal information), by a straightforward modification of a standard pump-probe setup. This is possible by employing a concept of compressive imaging denoted as the single-pixel camera. We exploited laser light coherence for generation of a series of random encoding patterns, which provide us with ability to computationally reconstruct an image from the knowledge of the total transmitted probe intensity. [1]

Functionality of our setup was demonstrated on several proof-of-principle measurements. Namely, we have carried out imaging of a tilted pump beam gradually exciting a homogeneous sample of CdSe quantum dots, imaging with a structures pump beam and imaging of transient dynamics on two structured semiconductor samples.

The experiment provides a simple tool for femtosecond ultrafast imaging, which can be used in a number of spectroscopic laboratories. The use of compressive sampling shortens the measurement time, thus reducing the requirements put on the laser, setup and sample stability. Moreover, under certain conditions the setup can be used for imaging of probe wavelengths in the infrared spectral region, where the 2D array sensors suitable for pump-probe measurements are not commonly available.

References

[1] Žídek, Karel, Ondřej Denk, and Jiří Hlubuček. "Lensless Photoluminescence Hyperspectral Camera Employing Random Speckle Patterns." *Scientific Reports* 7.1 (2017): 15309.

Packaging-induced stress analysis in high-power DFB laser diode at 975nm

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High-power (10W) spatially multi-mode GaAs-based pump lasers have been developed for more than 20 years addressing modern telecoms applications (975nm pumps for Yb, Er and Yb/Er doped fibers). There is also an increasing interest in the micro-machining, medical applications…. However, the momentum for increasing their performances is still present in particular to operate at the highest level of emission power and reject the Catastrophic Optical Damage (COD) level threshold as far as possible. It is well documented now to consider that failure due to a change in optical properties can arise from two main failure mechanisms both of them largely activated by temperature :

1. Increased optical loss due to a localized (at the facet) degradation of the active region (QW) itself which leads to optical absorption and/or scattering (Catastrophic Optical Mirror Damage COMD),
2. Bulk damage or defects of material around the active region, which absorbs and/or scatters light.

Catastrophic optical damage (COD) corresponds to a sudden degradation as failure mode that affects edge-emitting diode lasers in particular when operated at elevated levels of emission power. In most cases, COD starts at one of the cavity mirrors while sometimes defects within the gain medium inside the cavity also initiate the COD process. There are numerous reports on COD effects and kinetics, most of them relying on the analysis of the resulting damage pattern after COD has finished. For high-power broadband Laser diodes, works have been initiated since few years ago in particular by the research group of J. W. Tomm from the Max-Born Institute in Berlin. This Group has produced background and reference papers on experimental and theoretical study of temperature distributions inside the bulk and at the facets.
The development of 9XXnm pump Lasers has gone through several generations of devices to reach full maturity, high reliability and unprecedented output power levels in this decade by improving both facet robustness and reducing internal loss. Moroever optical fibers Yb, Er and Yb/Er have a narrow absorption line close to 975nm, hence the pump source needs a spectral width < 1nm. Multimode 975nm laser diodes are the best choice as a pump source and the requirement of wavelength stabilization with temperature is more obvious. Such narrow spectral width (<1nm), combined with wavelength thermal stabilization (0.07nm°C-1), is achievable in the high-power diode lasers using optical feedback provided by a uniform distributed feedback grating (DFB) introduced by etching and re-growth process techniques. This results in the development of DFB broad area (90µm wide) chip able to emit 10W @ 25°C. Nevertheless and although these huge improvements, COD is still a major concern thus limiting performances and operating reliability of such devices since the COD threshold is more and more rejected towards high current values and COD sensitivity is more and more difficult to predict on fresh devices.
Since several years, the Optoelectronic device group at MBI deals with the kinetics of COD. Main emphasis of the research is laid on the identification of the microscopic mechanisms that jumpstart sudden degradation. Their identification allows to understand self-reinforcing processes in all types of optoelectronic devices (and light guiding materials), independent of a particular device structure or architecture. For this purpose, it is crucial to have available test devices, which are superior in terms of homogeneity, reproducibility and performance. At the present stage, this is exclusively ensured in cooperation with world-leading industrial partners. The proposed cooperation between both the Photonic devices team of IMS-CNRS Lab in Bordeaux, the Photonics group of the 3-5 Lab and the Optoelectronic device group at MBI, exactly addresses the above synergy effects resulting from the inputs of the leading device producers and analysts.

The Chip on Submount (CoS) devices have been provided by 3-5 Lab according some specific designs. Specifically, the work that has been addressed at MBI facilities in the framework of the LASERLAB Europe program has been conducted on:

1. Time-resolved measurements using thermal streak-camera in pulsed regimes by increasing magnitude of pulses along the lateral side of the CoS Laser diodes.
2. Measurements of facet temperature by µ Raman and thermo-reflectance.
3. Pulsed COD measurement on different devices.

Stress distribution analysis as well as Catastrophic optical damage (COD) threshold determination has been achieved and discussed on high-power 975nm DFB LDs with different vertical symmetric and asymmetric structures using these three dedicated techniques. All these results have been recently included in a PhD study (R. Mostallino) that has been defended on September 2018 at the University of Bordeaux.

Sub-terahertz radiation from large aperture photoconductive antennas excited by UV laser beam

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ABSTRACT

Intense THz radiation sources have recently witnessed significant advances, opening the study of the nonlinear interaction between matter and THz waves, such as high-frequency generation and field-effect emission [1,2]. Large-aperture photoconductive antennas (LAPCA) are optically pumped sources of intense THz radiation [3], generating unique quasi-half-cycle THz pulses with low frequency components, the latter being advantageous when requiring high ponderomotive potential [4]. In this work, we study the generation of THz pulses from LAPCA that uses wide bandgap semiconductor crystals (ZnSe, GaN, 6H-SiC, 4H-SiC and β-Ga_2O_3), pumped by KrF laser pulses at 248 nm wavelength, with optical energies up to 80 mJ and pulse durations above 700 fs. We studied the scaling of the THz energy as a function of the laser energy and bias electric field, and found that 6H-SiC and 4H-SiC were the most efficient LAPCAs. According to autocorrelation measurements, the generated frequencies were in the sub-THz range, which is a direct consequence of the long optical pump pulse duration. Additionally, by performing Z-scan experiments, we demonstrated a 1.7-times non-linear transmission enhancement inside an n-doped InGaAs semiconductor using these intense sub-THz pulses.

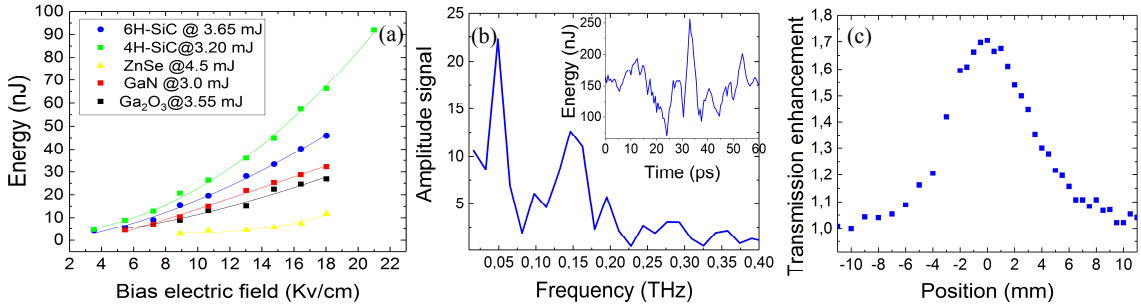

Fig. 1: (a) Scaling of the THz energy as a function of bias electric field for ZnSe, GaN, β-Ga_2O_3, 6H- and 4H-SiC LAPCAs. (b) THz spectrum of the THz pulse generated by the 4H-SiC interdigitated LAPCA. The inset shows the autocorrelation trace. (c) Normalized transmission of the Z-scan experiment from the n-doped InGaAs layer with the intense THz pulses radiated by the 4H-SiC interdigitated LAPCA.

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[2] G. Herink *et al.,* New J. Phys. "Field emission at terahertz frequencies: AC-tunneling and ultrafast carrier dynamics" **16**, 123005 (2014)
[3] X. Ropagnol *et al.*, Appl. Phys. Lett. "Intense terahertz generation at low frequencies using an interdigitated ZnSe large aperture photoconductive antenna" **103**, 161108 (2013)
[4] X. Ropagnol *et al.,* Opt. Express, "Intense THz pulses with large ponderonmotive potential generated from large aperture photoconductive antennas" **24**, 11299 (2016)

Highly resolved Rydberg-transitions in X-ray spectral range of vanadium ions in dense kJ laser produced plasmas at PALS

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The observation of X-ray spectra from Rydberg atomic states in dense plasmas opens unique possibilities for the determination of the transition between discrete and continuous spectra and the analysis of the impact of electric fields on atomic populations. In a kJ-ns laser produced plasma at PALS we have implemented a particular high-resolution spectroscopy in X-ray spectral range to study the impact of electric fields on atomic populations of the fine structure of resonance X-ray transitions.

Time-resolved study of the laser mediated interactions between specially designed model system peptides

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Name of the Laserlab access facility: SLIC

ABSTRACT. CrossLink (PI-CL) between proteins is a crucial tool in unveiling protein-protein interactions in various contexts. To evaluate the involvement of aromatic residues in the inter/intrasubunit CL, we have (i) studied the photo-induced dimerization of natural peptides such as Xenopsin, and (ii) performed systematic photophysical and photochemical studies of a series of small rationally designed peptides in order to understand the structural determinants of the PI-CL-induced bond formation.

As for (i) Xenopsin, that can be viewed as a simplified in-vitro model system for protein-protein interactions in some cases, contains a single aromatic amino-acid, Trp, which is an efficient absorber of UV (250-300 nm) light, thus capable to trigger PI-CL. The dynamics of the PI-CL was first investigated by Flourescence Up-Conversion by irradiating water-based solutions of Xenopsin with 260-nm, 250-fs laser pulses. In this experiment a main difficulty encountered consisted in the photo-damage induced to the initial monomeric species, even at the lowest possible irradiation power. Thus, we changed our strategy: we formed Xenopsin dimers first, by appropriate irradiation of the initial species with 260-nm fs pulses, and then we investigated the relaxation dynamics of the formed stable dimers by Time-Correlated-Single-Photon-Counting (TCSPC), by using pulses in the few kW/cm^2 peak intensity range, not to perturbe the species under study. Mass Spectrometry confirmed that the irradiated samples were enriched in dimer content due to UV laser irradiation. Formed dimers are responsible for a new absorption band of the irradiated target in the 350-400 nm window, whereas the monomer absorbs in the deep UV up to 300 nm.

As for (ii), we characterized first the simplest systems, represented by the free aromatic amino acids, Tyr and Trp, respectively. Their response was compared to that of the specially designed model peptides containing two aromatic residues (Tyr/Tyr, Tyr/Trp, Trp/Trp) in order to see if molecular organisation promotes intra-chain CL over inter-chain. Several small peptides, with Glycine linkers if different lengths, were investigated, having the general structure Tyr/Trp-(Glycine)n-Tyr/Trp with n=2,3,4, in order to shed light on the conformational requirements for laser-mediated cross-linking to occur. We performed fluorescence lifetimes measurements on three small peptide families with excitation wavelengths of 280 or 289 nm. Fluorescence decays were recorded at 310 nm (peak of Tyr emission) and 350 nm (peak of Trp emission). In addition, we made comparative measurements on monomeric Trp and Tyr. We carried on two types of measurements: (1) in a 3.5 ns time interval at parallel and perpendicular excitation/detection polarizations in order to evaluate the fluorescence anisotropy decays of the excited peptides and (2) in a longer 20 ns time interval at magic angle polarization in order to measure the lifetime precisely. We observed that In all cases, the fluorescence decays of the peptides are much faster than the Trp decay. A number of other interesting and somewhat unexpected results concerning the relaxation dynamics of the peptides was found, as some surprisingly short lifetime related to excited states of the molecular species which is apparently independent of the length of the Glycine linker.

Imaging nanoscale aggregation of proteins ex vivo using the contrast in Förster resonance energy transfer obtained from 2D polarization fluorescence imaging (2D POLIM)

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Förster resonance energy transfer (FRET) is a well-established nanoruler acting on a distance of 2-10 nm. It can be used to discriminate small aggregates of fluorescent molecules or nanocrystals from just high concentration without aggregation. One drawback of conventionally spectrally resolved FRET measurements is the need of a pair of different fluorescence labels. Polarization resolved fluorescence microscopy allows for the determination of the so-called *homo-FRET* between similar fluorophores. However, conventional fluorescence anisotropy measurements are restricted to the study of isotropic samples, as their results otherwise will depend on the choice of the lab frame. LLC provides the possibility of 2D polarization imaging (2D POLIM), which was recently developed by Ivan Scheblykin and coworkers.[1,2] 2D POLIM combines controlled variation of the orientation angles of linearly polarized excitation and of fluorescence detection, thus, providing full in-plane information of the polarization state of the sample.

2D POLIM has proven its power in materials research,[1–4] and was recently applied for a semi-quantitative investigation of early protein aggregation of GFP-labeled human α-synuclein in models of Parkinson's disease ex vivo.[5] Within the access project LLC002451 we applied 2D POLIM to the investigation of *apical f-actin aggregation in healthy and cholestatic liver tissue*, which is related to liver damage in severe sepsis.[6] A qualitative analysis of the measurements showed a local variation of the FRET parameter obtained from phalloidin-stained f-actin, which can be related to the pathologic condition of the sample.

References:

(1) Camacho, R.; Thomsson, D.; Yadav, D.; Scheblykin, I. G. I. Quantitative Characterization of Light-Harvesting Efficiency in Single Molecules and Nanoparticles by 2D Polarization Microscopy: Experimental and Theoretical Challenges. *Chem. Phys.* **2012**, *406*, 30–40.

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Regulation of root hair growth in the model plant Arabidopsis thaliana

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Plant growth and survival are dependent on the acquisition of water and nutrients by roots. To optimize resource capture in a heterozygous soil environment, root growth and morphology are constantly modified. One of the modifications is an alteration in the density and length of root hairs (RH), which are long finger-like extensions of specific epidermal cells that enlarge the root's absorptive surface. Changes in the abundance or activity of specific signalling molecules lie at the base of this root growth plasticity. Among the development and growth regulating molecules, the plant hormone auxin is recognised as the most influential. RH growth is dependent on vesicle delivery and fusion to the apical membrane, which constantly changes the overlying apical cell wall. In this work, we wanted to identify direct targets of auxin signalling in RHs and find a direct link between auxin and RH cell wall signalling, which was mostly lacking before.

Using a screen for aberrant RH morphology, we identified *ERULUS*, a member of an *Arabidopsis* receptor-like kinase family of putative cell wall sensor proteins. Mutant *erulus* RHs were short (22% of WT), bulged, grew irregular and 3-fold slower compared to WT RHs. Complementary sets of experiments illustrated that *ERULUS* is a direct target of auxin. Next, we investigated ERU function. Throughout RH development ERU-GFP localized to the apical plasma membrane, the site of active cell wall deposition/modification. As a result of the mutation in ERULUS, RHs exhibit severely altered dynamics of specific components (pectin) in the apical cell wall, and a continuously higher pectin Ca^{2+}-binding capacity, a *sine qua non* for cell wall cross-linking. This indicates that ERULUS is required in fine-tuning the balance between having an extensible, yet strong cell wall at the growing tip. Based on these results, we hypothesized that the aberrant temporal relationship between *erulus* growth and pectin Ca^{2+} binding site oscillations could lead to a mistiming/uncoupling of downstream oscillatory events, further exacerbating the *erulus* phenotype.

Since $[Ca^{2+}]_{cyt}$ oscillations are thought to follow growth rate oscillations, and specific $[Ca^{2+}]_{cyt}$ signatures control specific downstream events, we transformed a Förster Resonance Energy Transfer (FRET)-based calcium sensor (Yellow Cameleon 3.6) in *erulus* mutant plants and quantified growth/$[Ca^{2+}]_{cyt}$ interdependency using the Single Plane Illumination Microscopy (SPIM) setup at the CUSBO facility in Milan. Consistent with earlier reports on growth/calcium behavior in growing WT RHs, we confirmed that growth rate oscillations tend to anticipate $[Ca^{2+}]_{cyt}$oscillations by a few seconds. In *erulus* RHs however, we observed that (1) a wider range of phase delays is present with (2) growth often lagging behind $[Ca^{2+}]_{cyt}$ oscillations. Upon external calcium supplementation, part of the *erulus* RH phenotype was rescued. These findings suggest that loss of ERULUS function leads to a mistiming/uncoupling of growth rate and $[Ca^{2+}]_{cyt}$ oscillations, a perturbation which could affect downstream pathways crucial for sustained tip growth.

In summary, all data suggests that ERULUS acts to fine-tune growth at the apical RH tip.

Photoelectron circular dichroism of fenchone from multiphoton ionization with nanosecond laser pulses

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Photoelectron circular dichroism (PECD) is a highly sensitive enantiospecific spectroscopy for studying chiral molecules in the gas phase using either single-photon ionization (with synchrotron radiation) or multiphoton ionization (with tabletop femtosecond laser systems). In the short pulse limit investigated with femtosecond lasers, resonance enhanced multiphoton ionization (REMPI) is thought to be rather instantaneous, whereas on longer timescales other types of dynamics such as internal conversion or molecular rotation may play a role. As PECD is a sensitive probe of structural dynamics such as conformational isomerization, a growing interest in vibrational sensitivity of PECD has emerged. In the presented work we extend previous findings on vibrationally state-selective PECD measurements to track the influence of the vibrational level of the intermediate state populated in the 2+1 REMPI of fenchone. The results indicate no strong influence of vibrational level, to within experimental uncertainty (10% fluctuation). Our study also demonstrates the feasibility of PECD with nanosecond dye laser systems, which are significantly cheaper than the femtosecond tabletop systems used to date.

This study has been carried out in a collaboration of scientists from Göttingen University, Kassel University and scientists from the Institute of Electronic Structure and Laser at FORTH in Heraklion at the LASERLAB-EUROPE facilities in Heraklion.

ULTRAFAST STRUCTURAL DYNAMICS PROBED BY PHOTOELECTRON SPECTROSCOPY

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This work aims to study laser-induced lattice modifications in metallic samples at ultrafast timescales where there is a strong electron-lattice temperature non-equilibrium. This regime is difficult to model theoretically due to the limited knowledge on material properties[1], such as the electron-phonon coupling factor, the electronic and ionic thermal conductivities and the electron heat capacity. In this context, time resolved photoelectron spectroscopy (Tr-PES) experiments may help to improve our understanding of the physical processes and material properties in this regime. The excitation and relaxation of matter can be diagnosed by studying the temporal evolution of their PES spectra. However, such a pump/probe experiment, based on the detection of probe-induced photoelectron spectra is technically challenging due to the overlapping and interaction with the pump-induced high flux of photoelectrons[2].

For that purpose, we have developed a dedicated XUV beamline based on High-order Harmonic Generation that delivers femtosecond pulses with the required photon energy (80 - 100 eV) to partially overcome this issue while maintaining the sub-100 fs temporal resolution, essential for the study of this regime of interaction. This beamline has been installed at the CELIA laboratory on the Aurore laser facility and its characteristics will be fully described.

On the other hand, the mutual interaction of probe and pump-induced photoelectrons, called space charge effect, can distort the detected spectrum causing a loss of information. The control and understanding of this effect is thus of major importance for the development of such Tr-PES experiments. Our theoretical approach to describe this phenomenon will be presented together with our results to overcome this issue, as illustrated on the figure 1.

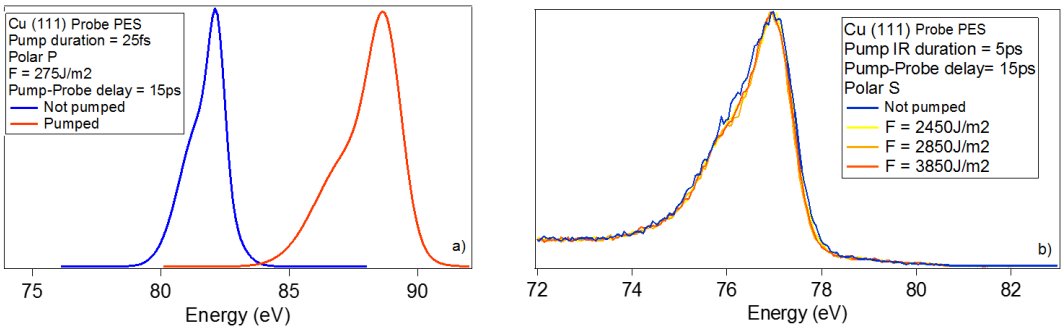

Figure 1 Pump-probe measurements of Cu PES corresponding to valence band a) shifted and broadened by space charge effect and b) without space charge effect by rotating the pump polarization and increasing the pump pulse duration.

Finally, the perspective of Tr-PES pump/probe experiments free of space charge effect where the lattice dynamics upon fs IR excitation could be observed will be discussed.

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Non-linear effect of the interaction of the self-trapped excitons under dense laser excitation in molybdates

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The interaction of self-trapped excitons (STEs) occurs in case of their high concentration in the local volume [1]. The effect is described as an Auger-like dipole-dipole interaction when the distance between STEs smaller than the radius of dipole-dipole interaction ($R_{d\text{-}d}$). Two experimental approaches were proposed recently for the study of the interaction of STEs. The method of the interband Z-scan luminescence yield implies the variation of the excitation density on the sample and allows to determine kinetic order of non-linear quenching of luminescence intensity, which provides with the information on the mobility of charge carriers in the crystal [2]. The study of the rate of non-exponential decay of STEs emission for variable excitation density allows to obtain the data on the dynamics of relaxation phenomena including the radius of dipole-dipole interaction of excitons [3,4]. Here we present the study of the STEs interaction in $ZnMoO_4$ and $CaMoO_4$ single crystals using the combination of these two approaches in order to obtain the most complete set of parameters of the phenomena. The studies were performed using the third harmonic of a Ti:sapphire laser at the CELIA laser center. Non-linear quenching of luminescence starts for concentration of excitations above $3*10^{20}$ for $CaMoO_4$ and $2.5*10^{21}$ cm^{-3} for $ZnMoO_4$. A prominent increase of the fast initial part of decay curve was detected with the increase of the excitation density. The numerical simulation of the measured decay and z-scan curves was performed with account for 3D nonuniform distribution of the initial concentration of excitations. The dipole-dipole interaction radii $R_{d\text{-}d}$ are calculated using the model which accounts for STEs diffusion and exciton thermal quenching.

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Ultrafast manipulations of high-energy spin excitations

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The investigation of the interaction between femtosecond laser pulses and magnetic materials has already revealed the tremendous potential of this approach for the ultrafast manipulation of spins [1,2]. In particular, the all-optical control of the magnetic order in antiferromagnets has become relevant, given the recent surge of interest in this class of materials for spintronics purposes. The peculiarity of laser pulses, when compared to other stimuli, consists in the possibility to excite, manipulate and detect spin excitations on the femtosecond timescale implying a direct access to the characteristic timescale of magnetic eigenmodes in antiferromagnets.

Recently the highest-frequency modes, which are magnons with wavevector near the edges of the Brillouin zone, have been impulsively photo-excited via a coherent light-scattering approach [3]. Remarkably, a complete manipulation of the phase and amplitude of coherent magnons with frequency equal to 22 THz and 1 nm wavelength (i.e. *femto-nanomagnons*) was achieved [3].

Even more exciting, further investigation of the femto-nanomagnonics regime demonstrated that it has little in common with the conventional spin dynamics triggered by nearly-zero-wavevector magnons. A novel theoretical quantum-mechanical framework has been developed, aimed at deriving a proper equation of motion was derived within our model; in addition an intruiging prediction suggests that the photo-generated pairs of magnons are quantum-mechanically entangled [4]. The elegant coherent Raman excitation does not allow a manipulation of the macroscopic phase of a magnetic material. A resonant excitation of high-energy magnons is expected to induce a strong perturbation of the magnetic system on the shortest time-scale. Recently we have explored the resonant excitation of a state of an electronic transition coupled to high-energy magnons. As a result of the direct injections of femto-nanomagnons, a magnetic phase transition is driven in less than one picosecond in a specimen of CuB_2O_4. Given the phase diagram of this compound, it follows that the phase transition implies a photo-induced magnetoelectricity which does not decay up to the nanosecond time-scale [5].

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Cooperative luminescence of Yb pairs in $Li_6Y(BO_3)_3$ single crystals

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$Li_6Y(BO_3)_3$ (LYB) may form solid solutions with homologous lithium rare earth (RE) borates. Therefore, LYB single crystals can accommodate large concentrations of such RE dopants like, e.g., Yb^{3+} ions. Crystal structure contains edge-sharing REO_8 polyhedra forming chains along the *c* axis allowing for closely situated RE^{3+} ions with a distance of about 3.85 Å. This geometry supports the emergence of cooperative luminescence (CL) that is the process in which two excited neighbouring RE^{3+} ions relax simultaneously emitting a single photon.

This report presents the results of spectroscopic study of the CL of Yb^{3+} pairs in LYB single crystals under continuous-wave laser excitation (Institute of Physics, Tartu) and pulsed excitation using a tuneable femtosecond laser system (Topas) at the Laser Research Center of Vilnius University, Lithuania. The CL peaking at about 500 nm was successfully generated in the temperature range of 6-300 K in LYB crystal doped either with 5 or 20 % Yb upon the excitation at the wavelength of 972.3 nm (Fig. 1a). The CL is nearly structureless at room temperature, but demonstrates a pronounced line spectrum at low temperatures. The main lines in the spectrum of pair centres can be described as a mere combination of the radiative transitions in single centres. The laser power dependence of the CL intensity was found to be quadratic at room temperature but nearly linear at 6 K. The decay kinetics of CL was twice as fast as that of a single ion luminescence and showed slight shortening with growing Yb concentration (Fig. 1b). The influence of temperature induced broadening of single centre absorption lines and energy transfer along the Yb chains on the power and temperature dependences of the CL will be discussed.

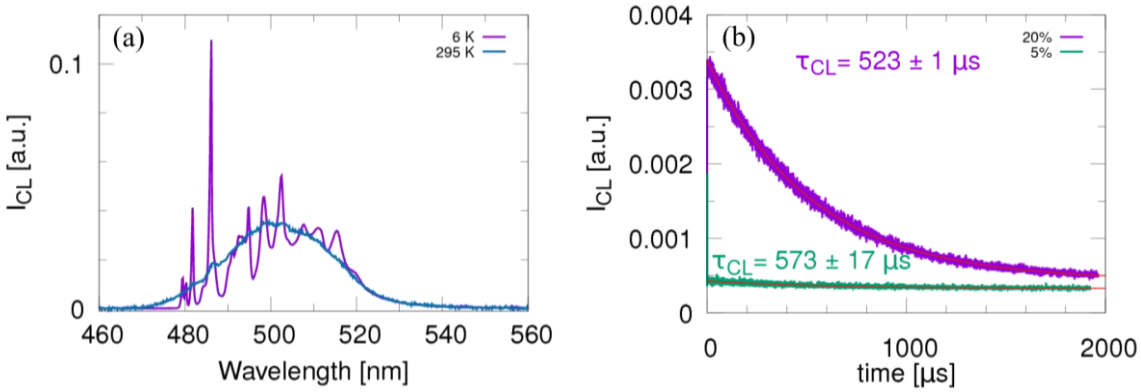

Fig. 1 CL spectra of the 20 mol% Yb doped LYB crystal measured at 6 K and 295 K (a). Evolution of the CL at 500 nm following the 40-ps laser pulse excitation at 972 nm in the 5 and 20 mol% Yb doped LYB crystals (b).

Sensitive Sub-Doppler Metrology in Cavity: from C_2H_2 to HD

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Despite its enormous success, the Standard Model of particle physics i.e., based on the Quantum Electrodynamics (QED), remains incomplete, e.g., it fails to explain the dark matter and the dark energy. On the other hand, current theories that attempt to unify gravity with the other fundamental forces predict variations of the fundamental constants. Variation of the spin structure coupling constant (α), implies violation of both Lorentz invariance, and of charge conjugation, parity, and time reversal (CPT). Another fundamental quantity of the Standard Model, the Proton-to-Electron Mass ration μ_{pe} needs to be tested for possible temporal drifts, while the proton size is still debated. Only tiny effects are anticipated which requires to select the most sensitive system. Rotational and vibrational molecular energies are quantities directly controlled by μ_{pe}. This requires extreme frequency resolution, i.e., the use of ultra-high resolution spectrometers, and of ultrasensitive detection techniques. In recent years, the test of QED has been applied to molecular hydrogen, the simplest objects available to test the fundamental physics concepts, or even their violation. Mass correction to the Born Oppenheimer approximation, including, adiabatic, nonadiabatic, relativistic and QED effects have been considered in the most accurate ab-initio calculations to obtain the energy levels of the Hydrogen Isotopologues. These calculations have revived the molecular hydrogen spectroscopy for probing fundamental physical constants. Any experimental derivation to the theoretical predictions can be interpreted in terms of possible New Physics features.

The molecular hydrogen dipole vibrational transitions are forbidden in the Born Oppenheimer Approximation, but mass correction adiabatic coupling partially allows these transitions in the heteronuclear systems. Nevertheless, these transitions remain weak. Hence, specific spectroscopy techniques need to be implemented to detect HD transitions in the fundamental vibrational and first overtone modes, for example.

The current Amsterdam project targets the first overtone ($v : 2 \leftarrow 0$) dipole transitions of HD in the Near InfraRed (NIR) at $\lambda \sim 1.38\,\mu$m. The Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectroscopy (NICE-OHMS) technique locked against a Cs-clock referenced Optical Frequency Comb (OFC) has been implemented. An optical cavity offers not only the unique advantage of enhancing the equivalent absorption length (monitored by the cavity finesse), but also the crucial capability of magnifying the intracavity electromagnetic field (EMF). Strong EMF can induce saturated absorption (Lamb dip resonance) of even weak transitions opening the door to a Doppler-free, i.e., to an extremely high precision spectroscopy.

The experimental sensitivity ($\sim 10^{-12}/\mathrm{cm}^{-1}/\mathrm{Hz}^{1/2}$) allowed us to recently determine the frequency center of 3 rotational transitions (R_1, R_2 and R_3) with an accuracy of the order of 30 kHz[1], i.e., with a gain of 3 orders of magnitude compared with the previous determinations.

However, the shape of the observed NICE-OHMS resonances suffer from unexpected abnormal asymmetric shape which may be attributed to the hyperfine structure of the rotational levels. Nevertheless, the shape of the NICE-OHMS dispersion resonances suggests possible interfering crossover resonances.

Hence, to understand the current HD results, we are exploring 2 main directions, i) the observations of additional HD transitions whose number of hyperfine components is reduced, ii) the study of benchmarking molecular species to validate the technique and to explore collision under low pressure regimes. Option i) requires a redesign of the current setup, option ii) addresses a few transitions of acetylene (C_2H_2) currently under study which will be presented.

Nonlinear spectroscopy modeling demands to accurately take into consideration, i) the possible interference between hyperfine transitions (crossover resonances), ii) the shape of the EMF, since under low pressure conditions, the transit time (or interacting time) become longer than the mean duration between 2 collisions. Intuitively, the transit time rate should limit the resonance width. However, resonances narrower than this rate have been observed. Hence, it is of crucial interest to validate our nonlinear absorption model dealing with Gaussian beam. This can be obtained by varying both, the pressure and the power conditions.

Actually, we think that the low pressure range (< 1 Pa) which can be explored with the current setup, is also an opportunity for studying the Velocity Changing Collision (VCC) under non-linear absorption conditions (shape and width of the sub-Doppler resonance), since the pressure broadening coefficient observed on various species, significantly differs from the values observed under standard Doppler-broadened conditions.

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Photoelectron circular dichroism and laser-induced electron diffraction of chiral fenchone molecules

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Photoelectron circular dichroism (PECD) is a fascinating phenomenon both from a fundamental science aspect but also due to its emerging role as a highly sensitive analytic tool for chiral recognition in the gas phase. PECD manifests in an asymmetry in the photoelectron angular distribution when a chiral molecule is ionized by circularly polarized light. Using femtosecond laser pulses, we have shown that a sensitivity on enantiomeric excess of better than 1% can be achieved [1]. Due to the small photon energies of optical light, femtosecond PECD usually proceeds through multi-photon ionization where intermediate resonances play an important role [2]. In this contribution, I will discuss the effect of pulse lengths on the PECD as well as on ionization yields through different intermediate resonances in the fenchone molecule.

In a different approach, we have started to perform laser-induced electron diffraction (LIED) on both enantiomers of fenchone. LIED images images the atomic structure of a molecule using an electron that was ejected from said molecule and accelerated in a strong laser field before rescattering on the molecule. For small molecules this methods allows for the determination of bond lengths and for the investigation of bond-breaking mechanisms [3, 4]. Here, we address the question if LIED – performed with linearly polarized light – can distinguish between the enantiomers of a chiral molecule.

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Dissociation of HCl at 0.4 K: Sequence Matters

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Aqueous acid dissociation reactions are at the heart of numerous chemical and biological processes. These reactions also play a significant role in atmospheric chemistry, most importantly, to the processes leading to ozone layer depletion. To understand the fundamental steps of these reactions, many theoretical and experimental studies have been carried out on small HCl-H_2O clusters, in the past. [1,2] In this study, we have studied the dissociation of HCl on stepwise addition of H_2O molecules in helium droplets. Ultrabright pulsed-free electron laser radiation at FELIX laboratory in Nijmegen, were used to measure the umbrella motion of the H_3O^+ moiety in the dissociated $H_3O^+(H_2O)_3Cl^-$ cluster, in the frequency range of 1000-1700 cm^{-1}. Using mass selective detection, we have unambiguously identified the spectral fingerprints of HCl dissociation taking place at 0.4 K. Our experiments along with high level ab initio MD simulations show that the dissociation takes place via a distinct reaction mechanism which is highly specific of the order in which molecular aggregation takes place. Details will be presented in the talk.

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Time resolved photoelectron circular dichroism @ FERMI

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A molecule is defined as chiral if it cannot be superimposed to its mirror image. The two mirror images of a chiral molecule are called enantiomers or optical isomers and they can be distinguished only when interacting with circularly polarized light sources or with another chiral molecule. Chirality is a very important property in biology and chemistry and even if it has been the subject of deep investigation, the ultrafast dynamics governing the chiral behaviour are still mostly unexplored.
The optimal investigation technique for the study of dynamics in chiral structures is the time-resolved photo-electron circular dichroism (TR-PECD), because of its strong sensitivity. In standard PECD, circularly polarized pulses ionize randomly oriented chiral molecules and the measurement of the angle-resolved photoemission presents a strong forward-backward asymmetry with respect to the propagation direction of the ionizing pulse. Recently, the first TR-PECD studies in chiral molecules have been realized, exploring PECD from electronic excited states and demonstrating the sensitivity of the technique as a probe of the ultrafast relaxation taking place in the chiral sample [1].
We wanted to extend this approach at the carbon K-edge to merge the chiral sensitivity of circularly polarized light with the chemical sensitivity of core excitation. Thus we exploited the unique properties of FERMI, as the only FEL providing circularly polarized XUV pulses up to the Carbon K edge with remarkable stability. In our experiment, we studied the ultrafast chiral dynamics occurring in fenchone when ultrafast linearly polarized pulses at 400 nm excite it by probing it with PECD at the carbon K edge (300 and 305 eV) by a circularly polarized FEL pulse.

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Measurements of fluence profile and plasma density in femtosecond laser filaments and superfilament in ambient air under different convergence of terawatt laser pulses

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The aim of the project was to investigate processes of multiple filamentation and superfilamentation [1] at multi-Terawatt laser radiation propagation in the ambient air. Different focusing conditions of Ti:Sa laser pulses with duration about 40 fs and energy of the individual laser pulse in range from 100 mJ to 130 mJ were under study. The reached intensity level and laser plasma density were measured.

By applying a new method for profiling intense laser beams suggested in [2], which is based on the dependence of the single-shot ablation threshold of common materials on the angle of incidence, the beam profile at a specific fluence level was straightforwardly recorded. The measurements show that in case of tight focusing (f-number about 15) the peak intensity, estimated from the measurements, exceeds 200 TW/cm^2 (the upper limit of the implemented technique). At the conditions which are very similar to the conditions in [1] it was shown that the peak intensity reaches 80 TW/cm^2 and the multiple filaments formed before the focal region without coalescing together into a single or a few superfilaments. The conclusions are supported by the numerical simulations [3].

The measurements of laser plasma density based on the method of precision interferometry [4] were performed at moderate (f-number about 50) and weak focusing with f-number 125. In first case the laser plasma at a single plasma channel formed during the separated formation of multiple filaments is about 4 times higher than in case of weak focusing, where the plasma channels with $0.5 \cdot 10^{17}$ cm^{-3} formed ~ 25 cm before the focal point was observed. The plasma density measurements confirm that multiple plasma filaments formed before the focal region does not form unit plasma channel.

Our experiments have been conducted using the JETI-40 laser system at the Institute of Optics and Quantum Electronics, the Friedrich-Schiller University in Jena, Germany. The research was supported by Laserlab-Europe Project (No. HIJ-FSU002344)

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Nanoscale lensless stereo imaging using dual coherent soft X-ray beams

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The ability to access 3D structural information of nanoscale objects is a challenging task and vital to various fields of scientific research. Common imaging techniques provide only two-dimensional projections along the imaging axis requiring, for a 3D reconstruction, several image recordings, extremely high radiation doses and long acquisition times. In particular, biological imaging entails non-reversible processes and can be highly sensitive to radiation damage, requiring single-shot experiments. We propose a technique, which allows retrieving three-dimensional amplitude and phase information from a single acquisition, by focusing two synchronized soft X-ray coherent beams with a calibrated stereo angle, on a nanoscale sample. From the two coherent diffractive imaging (CDI) reconstructions, a 3D reconstruction of the sample is achieved from the computation of quantitative disparity maps.

Schwarzschild objective alignment by means of wavefront diagnosis

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A self-designed soft x-ray Schwarzschild objective was realized for two purposes: i) Lithography imaging at-wavelength and ii) focusing of wide-angles soft x-ray sources in order to take advantage of the maximum throughput. The objective, based on B_4C/Ru normal incidence multilayer optics, was designed to compromise between optical resolution and modest high-NA aberrations. It provides a theoretical resolution of 60 nm and a magnification M=31X. However, in order to achieve such values the alignment of the optics as well as the finishing quality, are critical.

We evaluated the quality of the primary mirror by using a novel non-contact technique based on Hartmann-wavefront diagnosis [1]. The measurements were conducted at-wavelength, illuminating the multilayer coated mirror with the High Harmonic Generation Source of the Laboratoire d'Optique Appliqué (LASERLAB Host). The new optical surface metrology can resolve the surface of the mirror at nano-scale resolution. The results were compared with ray tracing and we concluded that the methodology is precise and easy to implement in other laboratories. Additional measurements will be conducted at the free-electron laser FLASH2 where the same technique will be used for the evaluation of the alignment of the optics.

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Single Shot XUV Nanoimaging Using an Intense Femtosecond Soft X-ray Laser

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Here, we investigate the wavefront properties of a high-harmonic-seeded SXRL depending on the time delay between the SXRL pump and HHG seed pulses. Specifically, we investigate the coherent wavefront directly at the output of the amplifying plasma channel for different time delays. For resolving the wavefront in amplitude and phase at the exit of the plasma channel, we image a micron scale target using ptychography and backpropagate the measured complex-valued illumination field to the exit of the plasma channel. Following characterization and optimization of the source properties, we successfully perform single shot nanoscale imaging.

The experiment was conducted at the Laboratoire d'Optique Appliquée with the 'Salle Jaune' Ti:Sapphire laser system pumping a SXRL using krypton gas producing Kr^{8+} ions in a 5 mm long plasma waveguide emitting at 32.8 nm. The high harmonic seed is derived from a gas cell filled with argon. The SXRL beam is separated from the optical beams by means of aluminum filters and reimaged using state-of-the-art curved multilayer mirrors (demagnification 1:10) onto a binary transmission sample consisting of a pattern of regular holes with 0.6 µm diameter and a spacing of 1.0 µm mounted in a x-y-z scanner. A CCD camera captures the coherently diffracted light downstream (numerical aperture about 0.25) and the sample was raster scanned over 30 scan points for ptychographic reconstruction by an implementation of the extended ptychographic iterative engine (ePIE) algorithm using position refinement. At each scan point single shot exposure of the sample by the HHG-seeded SXRL is performed. This allows reconstructing a high-resolution image of the sample and, at the same time, the complex-valued illumination field from the SXRL at the sample. We employ an aliasing free Fresnel propagator to propagate the field back to the exit of the laser plasma amplifier. This experiment is repeated for several time delays τ, where positive τ means the HHG seed pulse arrives following the SXRL pump pulse. For each delay step a full ptychographic reconstruction and backpropagation is performed, and, thus, allows quantifying the wavefront of the seeded SXRL out of the plasma channel in amplitude and phase. For a delay of 1.2 ps the highest photon flux is observed in the experiment, suggesting that highest gain is achieved in the plasma amplifier. The beam out of the laser plasma amplifier shows excellent spatial profiles. For increasing time delay, a decreasing mode field diameter is observed, while at the same time the wavefront curvature is reduced. Current investigations aim at unraveling the origin of the dynamics in the seed-plasma interaction. Following the characterization and optimization of the HHG-seeded SXRL we performed single-shot full-field coherent diffraction imaging. The recorded diffraction patterns are successfully reconstructed with a resolution in the order of ~100 nm at few hundred femtosecond exposure time.

In conclusion, we successfully employed ptychographic coherent diffraction imaging for characterizing the beam of a HHG-seeded SXRL operating at 32.8 nm wavelength in amplitude and phase with high fidelity. Backpropagation of the field allows determining source properties in unprecedented quality. We find that HHG seeding results in excellent spatial coherence properties, while a high degree of temporal coherence is maintained through the narrow-band amplification. Further, we find that the time delay dependence between the pump and seed pulses causes significant reshaping of the amplified laser beam hinting at a complex seed-plasma interaction that is subject of current investigations. Finally, we successfully employ the characterized and optimized HHG-seeded SXRL for single-shot coherent diffraction imaging demonstrating the outstanding capabilities for ultrafast nanoscale imaging.

Intense laser-energy coupling and partition in the transition to relativistically transparent laser-plasmas

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The absorption of laser energy into dense plasma and the resulting production of hot electrons underpins a host of intense laser-driven radiation production mechanisms. Using a suite of novel diagnostics developed for the PHELIX laser (GSI), we measure the total reflected laser energy as a function of intensity and demonstrate different absorption scalings for changing laser energy and focal spot size. The results of 2D particle-in-cell simulations, supported by an analytical model, indicate that the measured difference in scaling of absorption with focal spot size derives from changes to the way the refluxing electron population re-interacts with the incident laser pulse [1].

We then extend this study to investigate laser-energy coupling in targets undergoing so-called relativistic induced transparency. We find an optimum in the measured fractional absorption for targets on the order of 100's of nanometers and then a rapid decrease in absorption as the target thickness is reduced. Despite this reduction in total absorption, we measure a significantly higher temperature electron spectrum due to the re-heating of the escaping electron population by the transmitted laser. These insights may have important consequences for optimising laser-ion acceleration in the relativistically self-transparent regime.

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Quantitative X-ray Phase Contrast Imaging of a laser driven shock wave

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X-ray Phase Contrast Imaging (XPCI) [1] is a technique based on the photon phase-shift caused by an intense density gradient. It is therefore particularly indicated to probe materials which present density interphases such as a biological sample. However, this technique could also present several advantages compared to standard absorption radiography in the study of high energy density (HED) physics and warm dense matter (WDM). In particular, laser-induced shock-waves present high density gradients (in particular on the shock front) and they can propagate in materials at very different densities (e.g. multi-layer targets). To prove this, we performed an experiment at GSI using the laser PHELiX. In particular we used a ns laser pulse to lunch a shock-wave in a plastic cylinder and a sub ps laser pulse to generate a short X-ray back-lighter. The X-ray source was limited in space by the dimension of the target (5 μm diameter tungsten wire) to guarantee lateral coherence. From each experimental image, the amplitude and the phase map were extracted allowing a direct comparison with a hydrodynamic simulation, demonstrating the validity of such approach in HED and WDM physics.

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Enhanced relativistic-electron beam collimation using two consecutive laser pulses

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The double pulse approach to relativistic electron beam (REB) collimation has been investigated at the LULI-ELFIE facility. In this scheme, the magnetic field generated by the first laser-driven REB is used to guide a second delayed REB. We show how electron beam collimation can be controlled by properly adjusting laser parameters. By changing the ratio of focus size and the delay time of the two pulses we found a maximum of electron beam collimation clearly dependent on the focal spot size ratio of the two laser pulses and clearly related to magnetic field dynamic. Cu-K_α and CTR imaging diagnostics were implemented to evaluate the collimation effects on the respectively low energy ($\leq$ 100 keV) and high energy ($\geq$ MeV) components of the REB.

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