8th European Conference on Applications of Femtosecond Lasers in Materials Science

FemtoMat 2019

March 18 - 20, 2019
Mauterndorf Castle, Mauterndorf
Salzburg, Austria

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Chair: Wolfgang Kautek

Organization Committee: Oskar Armbruster, Aida Naghilou

Foreword

The science, technology, and application of femtosecond lasers matured to a level that a critical review of the state of the art is timely and possible to assist in signposting future trends of development.

The success of the series of the European Conferences on Applications of Femtosecond Lasers in Materials Science (FemtoMat) with

the 1st FemtoMat 2002, Visegrád, Hungary, October 2002,

the 2nd FemtoMat 2004, Bad Kleinkirchheim, Carinthia, Austria, February 2004,

the 3rd FemtoMat 2007, Vienna, Austria, April 2007, as special session of the 8th International Symposium on Laser Precision Microfabrication (LPM 2007),

the 4th FemtoMat 2011, Mauterndorf, Salzburg, Austria, March 2011,

the 5th FemtoMat 2013, Mauterndorf, Salzburg, Austria, March 2013,

the 6th FemtoMat 2015, Mauterndorf, Salzburg, Austria, March 2015

and the 7th FemtoMat 2017, Mauterndorf, Salzburg, Austria, March 2017, is the basis of the present

8th FemtoMat 2019, March 18 - 20, 2019, at Mauterndorf, Salzburg, followed by the Nano and Photonics Conference at the same venue.

The FemtoMat conferences represent a topical conference devoted exclusively to the present status and application of femtosecond lasers in materials science. The participants are encouraged to explore the possibilities and problems of the field and exemplify these by their most important new results. The conference features active discussions at the oral and poster sessions, and plenty of time for in-depth discussions in an informal atmosphere amidst the Austrian high alps. Due to the coherent topic of the meeting and to allow full attendance, no parallel sessions are being organized.

Vienna, March 2019

Wolfgang Kautek
(University of Vienna)

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Program

Monday, March 19, 2019

08:30 – 09:00 **Registration**
09:00 – 09:15 **Opening and greetings**
09:15 – 09:45 **L1** Leonid Zhigilei
University of Virginia, USA
"Material modification by laser-induced stresses and stress waves"
09:45 – 10:15 **L2** Tatiana E. Itina
LabHC, UMR CNRS 5516, F
"Laser structuring of porous nanocomposites: an interplay between photo- and thermo-activated processes"
10:45 – 11:45 **Posters:** Lectures and session

11:45 – 17:00 **Free discussion**

17:00 – 17:15 **Coffee**
17:15 – 17:45 **L3** Christian Spielmann
Friedrich-Schiller-Universität Jena, D
"Studying nonlinear absorption of intense laser pulses in a wide spectral range"
17:45 – 18:15 **L4** Thomas Winkler
University of Cambridge, UK
"Bringing nonlinear stimulated emission to the infrared: From Sapphire and fused silica to perovskites"
18:15 – 18:45 **L5** Aida Naghilou
University of Vienna, A
"Femtosecond laser generation of microbumps and nanojets on single and bilayer Cu/Ag thin films"
18:45 – 19:15 **L6** Alexander Bulgakov
HiLASE Centre, CZ
"Observation of repetition-rate dependent ionization of air by ultrashort laser pulses: Evidence for a role of metastable states of air molecules"

Tuesday, March 19, 2019

08:30 – 09:00 **L7** Gediminas Raciukaitis
Center for Physical Sciences and Technology, LIT
"Internal modification of fused silica with double femtosecond laser pulses"

09:00 – 09:30 **L8** Johannes Heitz
Johannes Kepler Universität Linz, A
"Micro/nanostructures from fs laser-processing for microneedles with controlled fluid transport and cell-repellent medical implants"

09:30 – 10:00 **L9** Cyril Mauclair
Laboratoire Hubert Curien St-Etienne, F
"Ultrafast laser surface structuring: the role of spatial beam control and in-situ characterization for process optimization"

10:00 – 10:30 **L10** Nadezhda Bulgakova
HiLASE Centre, CZ
"*Laser-matter interaction in the regimes of material processing: Toward more knowledge and better control"*

10:30 – 10:45 **Coffee**

10:45 – 11:15 **L11** Johannes Roth
University of Stuttgart, D
"New Developments in the Atomistic Simulation of Laser Ablation with Ultra-Short Pulses"

11:15 – 11:45 **L12** Klaus Sokolowski-Tinten
Universität Duisburg-Essen, D
"Non-equilibrium structural dynamics in nano-scale material systems"

11:45 – 17:00 **Free discussion**

17:00 – 17:15 **Coffee**

17:15 – 17:45 **L13** Peter Balling
Aarhus University, DK
"Ultrashort-pulse laser excitation of dielectric materials: What we think we understand and what we know we do not yet understand"

17:45 – 18:15 **L14** Eric Audouardr
AMPLITUDE, F
"Fs processing with high power laser"

18:15 – 18:45 **L15** Evgeny Gurevich
Ruhr-University Bochum, D
"Processing of semi-transparent materials with overlapping femtosecond laser pulses"

18:45 – 19:15 **L16** Thibault Genieys
LP3 Laboratory, F
"*Ultrashort laser excitation of Nickel for understanding energy deposition: Importance of non-thermalized electron distribution"*

Wednesday, March 20, 2019

08:30 – 09:00 **L17** Nicolas Sanner
Aix-Marseille University, F
"10-fs laser pulse ablation of dielectrics"

09:00 – 09:30 **L18** Linda Pabst
Laserinstitute Hochschule Mittweida, D
"Influence of pule duration on the selective ablation of aluminium thin films on silicon substrate using ultra short pulse laser radiation"

09:30 – 10:00 **L19** Walter Perrie
University of Liverpool, UK
"Femtosecond laser ablation of amorphous Polyethyl(ethyl)ketone (PEEK) at 775 nm and 387 nm"

10:00 – 10:30 **L20** Andreas Blumenstein
Laser-Laboratorium Göttingen e.V., D
"Gold Surface Nanostructuring with Ultrashort Laser Pulses - Study of Non-equilibrium Effects"

10:30 – 10:45 **Coffee**

10:45 – 11:15 **L21** Inam Mirza
HiLASE Centre, CZ
"Chemical functionalization of graphene on laser patterned surfaces"

11:15 – 11:45 **L22** Maximilian Spellauge
Munich University of Applied Sciences, D
"Contribution of pressure waves and material re-deposition to the energy specific ablation volume in femtosecond material processing with double pulses"

11:45 – 12:00 **Closing remarks**

Lecture Abstracts

L1

Material modification by laser-induced stresses and stress waves

M. Shugaev, M. He, L.V. Zhigilei

University of Virginia, Department of Materials Science and Engineering

Short pulse laser energy deposition leads to a rapid localized heating of the target material and, unavoidably, results in the generation of mechanical stresses. The laser-induced stresses can be grouped into three categories: (1) dynamic transient stresses and stress waves generated due to the condition of stress confinement, (2) long-term quasi-static thermo-elastic stresses related to the temperature gradients, and (3) residual stresses due to the laser-induced structural changes (defects) in the material. In this presentation, the mechanisms responsible for generation and relaxation of laser-induced stresses will be discussed. Several examples of material modification by laser-induced stresses and stress waves will be provided, including the generation of crystal defects within the laser spot and acoustic activation of surface processes at substantial distances away from the laser spot. The discussion of the mechanisms of responsible for the stress-induced material modification will be illustrated by results of large-scale atomistic simulations.

L2

Laser structuring of porous nanocomposites: an interplay between photo- and thermo-activated processes

H. Ma[1], N. Sharma[1], F. Vocanson[1], S. Bakhti, N. Destouches[1], D.S. Slaughter[2], Y. Andreeva[3], M. Sergeev[3], T. Itina[1,3]

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[2] Chemical Sciences, Lawrence Berkeley National Laboratory, Berkeley, USA
[3] ITMO University, Saint-Petersburg, Russia

Laser irradiation of nanoporous matrices doped with metallic saults is used for synthesis and structuring of several nanocomposite materials. Because of their unique properties, these materials have found many applications in optics, photonics, optoelectronics, security and medicine.

An efficient control over both laser fabrication and properties requires a better understanding of interplay between a set of photo- and thermo-induced strongly non-linear processes. In this talk, experimental findings will be analysed based on the developed self-consistent 2D models allowing us to connect the optimum laser parameters with the desired structural and optical properties.

L3

Studying nonlinear absorption of intense laser pulses in a wide spectral range

R. Hollinger[1,2], Z. Samosonva[1,2], D. Kartashov[1], S. Alisauskas[3], V. Shumakova[3], A. Pugzlys[3], A. Baltuska[3], R. Röder[4], C. Ronning[4], C. Spielmann[1,2]

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[2] Helmholtz Institute Jena, Jena, Germany
[3] Photonics Institute, TU Vienna, 1040 Vienna, Austria
[4] Institute of Solid State Physics, FSU Jena, 7743 Jena, Germany

Nonlinear optics with few-cycle laser pulses in nanoscale solids is of great importance for fundamental research and practical applications. Additionally interaction with nanostructured wide-gap semiconductor targets allows e.g. the observation of nanolasing under multi-photon pumping and enhanced optical frequency conversion. For all these experiments a precise determination of the damage threshold is crucial, because it limits the maximum intensity for nondestructive experiments. To estimate the amount of absorbed energy, we measure not the damage threshold; we measure the emitted fluorescence yield which is proportional to the amount of absorbed laser energy independently of the absorption mechanism. For this type of experiments nanostructured targets have another advantage, because they can form a laser cavity and we can observe lasing, and from the threshold we can directly estimate the number of excited carriers.

As we are mainly interested in damage-free interaction up to very high laser intensities, long wavelength femtosecond pulses are of great interest. Decreasing the photon energy into the mid-IR spectral range increases the multiphoton order which implies an increase of the damage threshold. However, at long wavelengths we observe the transition from multi-photon to tunneling excitation as the dominant absorption mechanism, which has been not studied for solids in great detail. Additionally at longer wavelengths we have to consider an enhanced avalanche ionization rate, which can lower the damage threshold. Here we present an extensive study of the nonlinear absorption of wide-gap semiconductor materials such as ZnO in a wide wavelength range from the visible to the mid-IR, by observing the emission as function of many parameters. As a further proof of the different absorption mechanism for different wavelengths we will present first results of the emission for different input polarization. These studies will help to better understand the nonlinear absorption, and the subsequent estimation of damage thresholds for different materials in wide range of parameters.

L4

Bringing nonlinear stimulated emission to the infrared: From Sapphire and fused silica to perovskites

T. Winkler[1], S. Bourelle[1], P. Balling[2], T. Baumert[3], F. Deschler[1]

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[2] Department of Physics and Astronomy, Aarhus University, Denmark
[3] Institute of Physics and CINSaT, University of Kassel, Germany

While the stimulated emission of light was postulated over 100 years ago and has found its way into nearly every laboratory, its nonlinear counterpart has only been observed in a handful of experiments so far. Therefore, it was very surprising, when we recently discovered the nonlinear coherent amplification of an ultraviolet femtosecond laser pulse in a piece of optically excited sapphire (LADIE effect [1]). The nonlinear (i.e. two-photon) stimulated emission holds high promises for laser technologies, microscopy or laser-spectroscopy as it is inherently nonlinear and provides a different set of selection rules. Here, we present extended studies showing the possibility of switching between two nonlinear amplification processes in fused silica, one being related to the LADIE effect, whereas the other is related to the characteristic and long-living self-trapped exciton states. Furthermore, we discuss our recent experimental studies to expand the nonlinear stimulated emission from the ultraviolet into the infrared regime. To that extend we utilize novel 2D and 3D perovskite materials, i.e. $MAPbBr_3$ and $BAPbI_4$, which showed already great properties for optoelectronic devices. Having band gaps in visible spectrum around 500 nm and long carrier lifetimes they are an ideal sample system to probe the two-photon stimulated emission in an ultrafast pump-probe experiment with 1000 nm infrared probe pulses.

[1] T. Winkler et al. Nature Physics volume 14, pages 74–79 (2018)

L5

Femtosecond laser generation of microbumps and nanojets on single and bilayer Cu/Ag thin films

A. Naghilou[1], M. He[2], J.S. Schubert[1], L.V. Zhigilei[1,2], W. Kautek[1]

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[2] University of Virginia, Department of Materials Science and Engineering, Charlottesville, Virginia, USA

Femtosecond laser ablation of metal thin films has been of vivid interest since decades. The generation of microbumps and nanojets have been observed and reproduced in many studies. It is generally accepted that frozen nanojets are produced through rapid melting. However, the processes responsible for the formation of microbumps, is still under discussion. In particular, subsurface boiling and pressure of the vapor released at the substrate-film interface, melting and redistribution of molten material, and plastic deformation of the irradiated film have been suggested as processes responsible for the formation and breaching of the microbumps.

In this study, the mechanisms responsible for the formation of microbumps and nanojets on Ag/Cu thin films and double layers deposited on a glass substrate and irradiated by a single 60 fs laser pulse are investigated experimentally and with atomistic simulations. The composition of the laser-modified bilayers is probed with the energy dispersive X-ray spectroscopy. The experimental observations are explained based on results of atomistic simulations, which reveal the important role of the difference in the electron-phonon coupling factor of the two metals in the mechanism of bump formation and breaching.

L6

Observation of repetition-rate dependent ionization of air by ultrashort laser pulses: Evidence for a role of metastable states of air molecules

A.V. Bulgakov, I. Mirza, V.P. Zhukov, N.M. Bulgakova, R. Machulka, O. Haderka, E.E.B. Campbell, T. Mocek

HiLASE Centre of the Institute of Physics CAS, Dolní Břežany, Czech Republic
Palacký University, Olomouc, Czech Republic
School of Chemistry, University of Edinburgh, UK

The vast majority of scientific and technological applications of short and ultrashort laser processing of materials are performed in a gas environment, mainly in air under atmospheric conditions. However, the role of ambient gases is so far insufficiently understood due to the complexity of laser beam propagation in ionizable media and the large variety of involved processes. For instance, high-repetition rate lasers, often used to increase the processing efficiency, can result in accumulation of the laser energy in the ambient gas if the repetition rate exceeds the rates of heat or excitation dissipation. While various accumulation effects, such as laser-induced formation of defect states and morphological changes on material surfaces and in volume, have widely been studied [1-3], little attention is paid to the fact that excited molecular and atomic states and hydrodynamic perturbations in ambient gases can also be accumulated, thus facilitating gas ionization by subsequent laser pulses.

In this talk, we report recent transmission measurements for femtosecond laser pulses focused in air at various pressures [4]. A spectral analysis of emission from the focal region has been also carried out. The air breakdown threshold and pulse attenuation due to plasma absorption are evaluated and compared with calculations based on the multiphoton ionization model. The plasma absorption is found to depend on the pulse repetition rate and is considerably stronger at 1 kHz than at 1-10 Hz. This suggests that accumulation of metastable states of air molecules plays an important role in initiation of air breakdown, enhancing the ionization efficiency at high repetition rates. Possible channels of metastable-state-assisted air ionization and the role of the observed accumulation effect in laser material processing are discussed.

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L7

Internal modification of fused silica with double femtosecond laser pulses

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Several approaches exist to induce the internal modifications in fused silica by femtosecond laser irradiation depending on the dose: direct writing of refractive index change (type I modification), birefringence control by nanogratings for geometric phase elements and polarisation sensitive imaging (type II modification) and new phenomena arising from double pulse utilisation.

Most of the studies show that the orientation of the LIPSS is perpendicular to the first pulse polarisation. However, the intra-volume modifications with the induced nanogratings have the depth dimension where the double-pulse fabrication can provide more sophisticated morphology depending on the temporal delay and energy relation between two pulses.

The nanogratings induced using the double-pulse irradiation with perpendicular polarisations demonstrates the grid-like structure at ~ 10 ps temporal delay, while the 45 degrees tilted gratings appear without delay between pulses. Variation of the nanograting period was observed in the case of parallel polarisation. Those new phenomena can be widely used for writing the two-dimensional diffraction gratings or the information coding applications and requires more deep investigations. A significant change in birefringence was monitored by varying the pulse delay in the range +/- 1 ps. Effects responsible for observed phenomena are discussed.

L8

Micro/nanostructures from fs laser-processing for microneedles with controlled fluid transport and cell-repellent medical implants

J. Heitz[1], C. Plamadeala[1], M. Muck[1], P. Fosodeder[1], A. Weth[2], W. Baumgartner[2], A.W. Hassel[3], C. Steinwender[4], J. Bonse[5], B. Buchroithner[6], J. Jacak[6]

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[6] University of Applied Sciences (Upper Austria), Linz, Austria

Laser-induced microstructures (written by two-photon polymerization using a Ti:sapphire fs-laser) can be employed to mimic features found in nature for controlled wetting or liquid transport [1]. Similar structures can be implemented into microfluidic devices to control the transport of fluids. We were able to produce biomedical microneedles covered with microstructures which induce the transport of an aqueous liquid to the needle tip [2].

Ti:sapphire fs-laser irradiation at a fluence above the ablation threshold can result in the formation of self-organized micro- and nanostructures, i.e., sharp cones or spikes covered by fine sub-wavelength ripples, at the surface of cylindrical Ti samples. The motivation of this part of our work is to create a structured ring on a small cylinder with cell-repellent properties [3,4]. We aim to create such structures on a miniaturized implantable pacemaker to avoid tissue growth around the device.

Acknowledgements:
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L9

Ultrafast laser surface structuring: the role of spatial beam control and in-situ characterization for process optimization

C. Mauclair

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Ultrafast laser pulses can be used to achieve structuring at the micro-nanometer scale by direct irradiation of the sample surface. The technique is gaining interest as nowadays ultrafast laser sources performances are more and more compatible with industrial requirements, and various surface functions can be achieved with this single step treatment. Thus, there is an interest in any technique that can reduce the laser surface processing time. In this contribution, some of the most efficient methods to take full advantage of the ultrafast laser sources are presented and discussed with a particular focus on spatial beam shaping and laser-scanner synchronization. Micrometric and sub-micrometric surface structuring can be achieved under certain irradiation conditions. Noteworthy, Laser Induced Periodic Surface Structures (LIPSS) can also be formed. The LIPSS dimensions generally range from 100 nm to a few micrometers. Thus, a thorough characterization of the irradiation result requires an additional step generally conducted after the laser irradiation by systems such as SEM and/or AFM with a resolution beyond the diffraction limit. Here, a super resolution microscopy technique based on structured illumination is used for in-situ observation of the irradiated surface. The technique permits to observe a phase locking mechanism during the multipulse formation of LIPSS, paving the way to real-time control of the ultrafast irradiation outcome.

L10

Laser-matter interaction in the regimes of material processing: Toward more knowledge and better control

N.M. Bulgakova, I. Mirza, V.P. Zhukov, T.J.-Y. Derrien, Y. Levy, A.V. Bulgakov, T. Mocek

HiLASE Centre of the Institute of Physics CAS

Development of new types of lasers with broad irradiation parameters widens the spectrum of viable laser applications in various fields of science, industry, and medicine. One of strategic goals of the HiLASE Centre is to develop novel applications of these versatile laser sources in material science. A laser application laboratory, which is now under completion, will combine state-of-the-art lasers with the most advanced diagnostics for investigations of interaction of laser light with different materials at the frontier of material science. An essential feature of the new application laboratory is a tight link between experimental and theoretical studies, a concept which already brings fruitful outcomes. Deep understanding of the fundamental phenomena involved in the laser action on materials of different kinds makes possible the elaboration of new approaches, new methodologies, and novel applications.

In this talk, the latest achievements of the HiLASE Centre in the field of laser material science will be overviewed. This includes comprehensive studies of ultrashort pulse laser action on transparent materials with new insights into mechanisms of material damage, revealing the ways for strong localized absorption of laser light inside bulk dielectrics with generation of pressures of hundreds of GPa, new understanding of the mechanisms of periodic nanostructuring of metals, and other latest findings. Several comprehensive theoretical approaches used in our studies will be unveiled, including one of the most advanced model for simulations of laser energy coupling to transparent materials under the regimes of direct laser writing of 3D photonic structures, with examples of their applications to real experimental conditions. Finally, further perspectives of laser-matter interaction studies at the HiLASE Centre will be discussed.

L11

New Developments in the Atomistic Simulation of Laser Ablation with Ultra-Short Pulses

J. Roth, E. Eisfeld, D. Klein

University of Stuttgart

The atomistic molecular dynamics (MD) simulation method has been combined with the two-temperature model (TTM) to successfully study laser ablation of metals, alloys and semiconductors with femto-second pulses.

Many electron-related parameters were kept constant in the original versions of combined simulation approach. We will present new developments for metals where heat conductivity and electron-phonon coupling are treated as functions of the electron temperature and absorption is calculated as a local function. For semiconductors the most important absorption mechanisms are modelled, the density of the electrons is treated explicitly and the interaction is modified as a function of the electron-temperature. Results will be presented for double pulses applied to metals and silicon. Further results include Al-Ni alloys and Al-Ni multi-layer systems.

The MD+TTM does not describe ions and free electrons explicitly. To be able to study the evolution of the plasma in the ablation plume an ionization model has been implemented and an interface has been created for mesoscale simulations of the plume.

L12

Non-equilibrium structural dynamics in nano-scale material systems

K. Sokolowski-Tinten

Faculty of Physics and Center for Nanointegration (CENIDE), University of Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany

We have carried out time-resolved transmission electron diffraction experiments at the MeV Ultrafast Electron Diffraction (UED) at SLAC National Accelerator Laboratory [1] to study non-equilibrium energy relaxation and transport in nanoscale materials after fs laser excitation. Precise measurements of the transient Debye-Waller-effect in different metal-insulator and metal-metal-heterostructures highlight the importance of interface effects [2,3] and reveal the competition between local relaxation and transport. At stronger excitation levels phase transitions may occur. Experiments on Au give direct evidence for a transition from heterogeneous melting at lower fluences to a homogeneous process at higher fluences [4].

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L13

Ultrashort-pulse laser excitation of dielectric materials: What we think we understand and what we know we do not yet understand

P. Balling

Dept. of Physics and Astronomy, Aarhus University, Ny Munkegade 120, DK-8000 Aarhus C, Denmark

The excitation of dielectric materials by ultrashort laser pulses is a multi-stage process, which involves strong-field excitation of electrons from the valence to conduction band, heating of the conduction-band electrons by inverse bremsstrahlung and potentially collisionally induced carrier excitation. The excitation is associated with fast and major changes in the optical properties of the material, which can be investigated experimentally in optical pump-probe experiments. Many of the current observations are explained by the general mechanisms outlined above, and in some cases, a quantitative agreement between measured data and models of the excitation process have even been obtained. The so-called multiple-rate-equation model has proven particularly useful for translating the mechanismic description into quantitative predictions of the dynamic material properties that are gauged in experiments. There are, however, still many unknown details. For example, a recent investigation by Winkler et al. [Nature Physics 14, 74–79 (2018)] demonstrated the surprising appearance of optical gain in a highly excited dielectric. In this presentation, I will focus on new, unpublished results from time-resolved pump-probe experiments with an 800 nm pump and a tunable probe measuring the time-dependent reflectance off normal incidence for both polarizations. The observed reflectances s- and p-polarized light are incompatible with a description based on a single value for the (complex) refractive index. Different explanations for this apparent discrepancy will be discussed.

L14

Fs processing with high power laser

E. Audouard, G. Bonamis, M. Delaigue, C. Hönninger, E. Mottay

AMPLITUDE, Pessac, France

Translating the high available power for industrial lasers [1] in the 100 W-300 W range into high throughput micro-processing is of high importance for future industrial applications. New basic questions are also raised using high repetition rates or high energies for ablation efficiency [2].
From the user perspective, dealing efficiently with high average power levels and high pulse repetition rates, e.g. in high speed scanners, requires an increase in the pulse modulation speed as well as free triggering in order to synchronize the laser pulses with scanner or axes positioning, and ultimately with the application. The challenge in femtosecond lasers is then to maintain the inversion level, and hence the output pulse energy, constant for any user profile. Thus, we can adapt the laser frequency to the variation of speed in case of complex movements, in order to maintain a constant fluence on the sample.

While ultrafast lasers excel processing with an exquisite precision, each laser pulse only removes a small amount of material. This disadvantage can be counterbalanced by distributing the energy into a burst of pulses at a GHz-level repetition rate [3,4]. Indeed, we conceptually show that GHz ablation efficiency is achieved through the balance of the pulses dedicated to heat accumulation and effective ablation. For that, several inter-related parameters, such as pulse energy, number of pulses per burst, intra-burst and inter-burst repetition rates must be optimized.

Beam divisions into multi beams using diffractive optics is also a development of particular interest. Programmable Spatial Light Modulators (SLM) can bring flexibility while maintaining a high spatial resolution compatible with complex multi spots shaping or user defined beam profiles. The recent technological progress in LCOS based systems, enables high optical transmission greater than 95 %, but also high average power handling up to at least 100 W of laser power. However, the dependence of the shaping performances on laser bandwidth places specific requirements when using ultrafast lasers. The usable field size with respect to the chromatic effects is for instance a specific parameter to manage. Optimization of the machining results by software driving of the SLM phase map can provide original and efficient solutions.
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L15

Processing of semi-transparent materials with overlapping femtosecond laser pulses

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We report on the experimental observation of selective delamination and recrystallization of semi-transparent materials upon femtosecond laser processing. Analysis of the delaminated layer indicates that the material undergoes melting on its both surfaces. The mechanism of delamination is identified as a complex interplay between the optical response of laser-generated free-electron plasma and nonlinear effects upon laser beam propagation in semi-transparent ceramics. Heat accumulation is shown to play key role for laser-induced recrystallization of amorphous metal oxides.

L16

Ultrashort laser excitation of Nickel for understanding energy deposition: Importance of non-thermalized electron distribution

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We present an experimental study and a theoretical analysis of femtosecond laser interaction with a Nickel sample. The evolution of reflectivity of near-infrared laser with pulse duration ranging from 15 fs to 100 fs is measured allowing us to have access to the deposited energy in the material. We provide a precise determination of ablation threshold fluence using the diameter-regression technique, a constant value of 0.33 +/- 0.025 J/cm^2 being found for the different pulse durations. We use a theoretical model to describe electron and lattice heating and their influence on the evolution of the reflectivity. Experimental results are compared to simulations based on the two-temperature model (TTM) and on a revised TTM taking into account the influence of electrons out of thermal equilibrium. This comparison highlights, here for Nickel, the importance of taking into account the nonthermalized electron distribution in the theoretical calculation of energy deposition excited by ultrashort pulses.

L17

10-fs laser pulse ablation of dielectrics

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The specific regime of few-cycle pulse interaction with dielectrics is investigated. Reaching the ablation regime requires only microjoules of energy. However, such conditions often require vacuum environment for proper propagation and focusing, because working in air ambience rapidly yields significant nonlinear spatial distortion of the beam in the pre-focal region, or even air ionization, which inevitably hampers the applicability of such ultrashort pulse. We first analyze the beam characteristics and discuss the inherent limitations to ablation of the surface of dielectrics when using a 10-fs pulse. Then we show the existence of an energy regime, above the ablation threshold, where the beam is still unaffected, authorizing proper ablation in air. We also explore the influence of the laser-created free-electron plasma at the surface of the material, and its reflective properties at the timescale of the pulse. By comparing fused silica and sapphire, we show that the critical plasma density is not systematically reached, even above ablation threshold fluences.

L18

Influence of pule duration on the selective ablation of aluminium thin films on silicon substrate using ultra short pulse laser radiation

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In recent years, the interest in selective structuring of thin films on different substrates played an increased role. There, the substrate material can influences the ablation process significantly.

The selective structuring of 30 nm thin aluminum films on silicon wafers with a 100 nm silicon oxide layer was investigated using ultrashort pulsed laser radiation (l = 1028 nm, tp = 0.2 – 10 ps, w86 = 15 µm). The influence of the processing parameters such as fluence, pulse number, scan speed and pulse duration was investigated.

Single and multiple pulse ablation thresholds were determined and the incubation coefficient was calculated. A different behavior in dependence on the pulse duration could be observed between the aluminum thin film and the substrate material. With increasing pulse duration the ablation threshold of the 30 nm aluminum thin film decreased, whereas, the ablation threshold of the silicon oxide increased. However, no significant influence of the incubation coefficient could be determined in dependence of the pulse duration. Therefore, the processing window for completely ablation of the 30 nm aluminum film without damaging the underlying silicon oxide increased with increasing pulse duration.

Furthermore, these results were applied to the selective structuring of the aluminum thin film using different scan speeds. With pulse durations in the femtosecond range a selective structuring could not be obtained at all investigated processing parameters. The substrate material was damaged prior to the complementally ablation of the thin aluminum film. With pulse durations in the lower picosecond range a selective ablation of the 30 nm thin film could be achieved without damaging the underlying substrate. The best results were obtained with pulse durations between 5 and 10 ps.

The processing window for a selective ablation of thin films on absorbing substrates was comparative small compared to none absorbing substrates, like glass. Due to the ablation thresholds were in the same magnitude for the thin aluminum film and the substrate material. A suitable selection of the pulse duration could significantly increase the processing window. Therefore, the pulse duration was an important processing parameter not only influencing the ablation quality but also to achieve a selective ablation of the thin film.

L19

Femtosecond laser ablation of amorphous Polyethyl(ethyl)ketone (PEEK) at 775 nm and 387 nm

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Laser micro-machining of amorphous PEEK has been demonstrated with 180 fs/1 kHz NIR (775 nm) and NUV (387 nm) laser pulses. The single pulse ablation threshold was found to be 2.01 ± 0.05 and 0.23 ± 0.02 J/cm^2 at 775 nm and 387 nm respectively. The significant difference in ablation threshold is due to the requirement for multi-photon absorption at 775 nm, where PEEK is transparent while linear absorption also occurs at 387 nm near the material bandgap. The incubation coefficients with multi-pulse excitation yield S(775) = 0.72 ± 0.01 and S(387) = 0.85 ± 0.02 respectively. Ablation of PEEK with NUV fs pulses shows much reduced melting and re-deposition, hence, precision NUV polymer micromachining is demonstrated. Laser induced periodic surface structures with pitch < 0.4 µm are observed at the base of ablated regions with 387 nm exposure. With the aid of a phase only spatial light modulator, multi-beam NUV micro-structuring is achieved, speeding micro-processing while reaching a line width < 4 µm using a high NA = 0.4 objective.

L20

Gold Surface Nanostructuring with Ultrashort Laser Pulses - Study of Non-equilibrium Effects

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Laser material interaction and structure formation in metals under controlled ultrashort laser pulse irradiation will be discussed. A novel approach is presented describing the exact amount of energy deposited by a laser pulse in a gold surface, applicable under a variety of structuring conditions. The presented model is validated by a newly introduced broad-band pump-probe reflectivity measurement method. The technique is also applicable to the investigation of other phenomena where a temporal resolution below 20 fs combined with an octave spanning spectral range is required. Here the method is used to probe the dynamics of energy deposition in a surface, which determines the evolution of the material status in the next few ps and ns, in which melting, ablation and finally crystallization in its final observable structure occurs. The influence of these laser induced phenomena on the formation of periodic nanostructures is investigated in the second part of the talk. The experimental results obtained in this work are compared to a large-scale molecular dynamics simulation. The novelty of this approach is the scale which the model calculations and the experiment covers, combined with the microscopic description of the electrons around the core with the macroscopic effects which an overheated melt has on the structure formation on the µm-scale. The interpretation of the obtained results leads to a deeper understanding of the involved process during the interaction of highly energetic ultrashort pulses with metals.

L21

Chemical functionalization of graphene on laser patterned surfaces

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Tuning of graphene's properties is the cornerstone towards reaching its ultimate potential. Recent advances have shown that periodic modulation of the pristine characteristics, such as potential and/or chemical functionalization, can unlock otherwise unavailable features. The reactivity of monolayer graphen also depends on the subtle substrate morphology, so nanopatterned substrates may provide a control over periodic modulation of chemical and physical properties of 2D-materials. Laser-induced periodic surface structures (LIPSS) is a universal phenomenon which can be observed on a variety of materials. Their properties depend on both materials properties and on the irradiation conditions. Here we report the results of a detailed study of micro-Raman microscopy on chemically functionalized graphene transferred on SiO_2/Si substrates covered with LIPSS. Periodic surface structures were fabricated using laser scanning method by systematically varying the laser energy fluencies and the spatial overlap of the focused Gaussian beam. The irradiation was performed on the bulk target using pulsed laser operating at central wavelength of 1064 nm with a pulse duration of 10 ps. The CVD–grown monolayer graphene was transferred to LIPSS-covered SiO_2/Si samples using the polymer support method. Results show that periodicity and depth of LIPSS influences the topography of the transferred graphene monolayer by either replicating the shape of the LIPSS, or by forming a periodic suspended pattern. The periodic modification of both mechanical and electronic properties of the monolayer graphene were studied. The optimum LIPSS periodicity-to-depth ratio, stress-strain analysis and functionalization efficiencies for the suspended and supported regions of graphene will be discussed.

Poster Abstracts

P1

Single- and Multicomponent Silicon-Based Nanoparticles due to Femtosecond Laser Nanostructuring and Their Applications

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Femtosecond laser nanostructuring is a versatile tool allowing fast synthesis of nanoparticles without any chemical precursors. They can be easily formed due to either femtosecond (fs) laser ablation of a solid target immersed in a liquid or laser decomposition of a micro- or nano-powder dispersed in a liquid. Variation of numerous experimental parameters leads to extensive change of size as well as structural and optoelectronic properties of formed nanoparticles. Moreover, multicomponent (bimetallic or semiconductor-metallic) nanoparticles can be easily formed by femtosecond laser treatment too. They can reveal widely variable properties as well as combine different functionalities in one nanoparticle depending on their structural properties.

Silicon nanoparticles are one of the most promising nanoelements that are found to be very perspective for biomedical applications due to their unique optoelectronic properties, biocompatibility and biodegradability. Femtosecond laser nanostructuring allows formation of contamination-free silicon nanoparticles of given size depending on a required application. In addition, their functionality can be also extended in consequence of additional femtosecond treatment resulting to plasmon properties of silicon-based nanoparticles. It leads to new potential applications of multicomponent silicon-based nanoparticles where initial ones cannot be employed. For instance, molecule detection using surface-enhanced Raman scattering by plasmon silicon-based nanoparticles is achieved recently.

In summary, femtosecond laser nanostructuring can be used for synthesis of both single- and multicomponent functional nanoparticles suitable for biomedical applications.

P2

Periodic surface structuring of fused silica, ULE glass and silicon using ultrashort laser pulses

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Femtosecond (fs) lasers have proven to be an excellent tool for processing of glass materials due to a reduced heat affected zone [1,2], as compared to ps and ns pulsed lasers. Besides surface ablation, micromachining and volumetric modification, fs-lasers have also appeared to be efficiently applicable in fabrication of various kind of laser induced periodic surface structure (LIPSS) [3]. For applications it is desirable to have LIPSS produced on large surface area with controlled periodicity. In this work, we will present results of a systematic study with the aim of producing LIPSS on fused silica (FS) and Ultra-Low Expansion (ULE) glasses. The irradiation of glass materials was performed using near-IR fs-laser in the fluence range close and well above the ablation thresholds of these two materials. We investigated the morphology of LIPSS formed in single- and multi-pulse regimes for both fixed irradiation spots and upon scanning the sample in a raster fashion under linearly polarized focused laser beam.

Upon scanning irradiation, in the case of ULE glass, coarse LIPSS with periodicity close to the laser wavelength and orientation predominantly perpendicular to the polarization of the incident laser beam were formed. For high-purity FS at the same irradiation conditions, a more complicated pattern of LIPSS is produced. Local regions with both perpendicular and parallel orientation and even the coexistence of the two orientations are observed.

Although nanostructuring by LIPSS is dominated by fs near-IR laser sources, increasing power of mid-IR picosecond laser sources enable to reach intensities sufficient for the modification of semiconductors and even glasses under focused laser beam. Possibility of using such laser sources for nanopatterning with periodic surface structures was tested on silicon.

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P3

Reaching the predictive stage in computing the energy absorbed by Si upon femtosecond laser irradiation in modification regime

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During the last decades, intensive studies of interaction of ultrafast laser radiation with materials have led to emergence of various applications based on functionalization of surfaces at the nano- and microscale. By inducing periodic modifications of material surfaces (band gap modification, nanostructure formation, crystallization or amorphization), optical and mechanical properties can be tailored, thus turning femtosecond lasers to one of the key technological tools for rapid development of nanophotonics, bionanoengineering, and nanomechanics fields.

Although the interaction of femtosecond laser pulses with semiconductor surfaces has been studied for more than two decades, the dynamics of coupling of intense laser light with excited matter remains far from complete understanding. In particular, swift formation of a transient overdense electron-hole plasma dynamically modifies optical properties in the surface layer of irradiated semiconductors and induces large gradients of hot charge carriers, resulting in ultrafast charge-transport phenomena.

In this work, the dynamics of ultrafast laser excitation of semiconductor materials are studied theoretically on the example of silicon. A special attention is paid to the electron-hole pair dynamics, taking into account ambipolar drift effects, the temperature-induced diffusive flows and the screening of the electron-phonon coupling. Modeling data on optical response has reached an excellent agreement with pump-probe reflectivity measurements available in literature [1]. Based on the simulation results, ultrafast processes such as non-thermal melting, scattering of light on the homogeneously nucleating liquid, and screening of the electron-phonon coupling are analyzed.

Achieving a predictive stage in describing the effects of intense laser light interaction with semiconductors enables the preparation of accurate modeling tools for the laser processing community [2,3,4].

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P4

Femtosecond and picosecond laser-induced damage thresholds of Si and Au in air and water

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Pulsed laser ablation in liquids (PLAL) is an efficient and flexible technique for nanoparticle production and surface nanostructuring. Although PLAL is simple in realization, the process itself is very complicated and still poorly understood. The complexity of the PLAL process can be illustrated by the example of the laser-induced damage threshold (DT). The DT is a well-defined parameter, can be quite reliably and unequivocally measured and provides a good reference for understanding and modelling of the physical process. However, the available data on DTs in liquids, when they are compared with DTs in air, are rather contradictory and various mechanisms such as light scattering [1], heat transfer to liquid [2, 3], reflectivity [4, 5] or vapor pressure [6] are used to explain the difference.

In recent years, several studies in nanosecond regime have been performed to determine DT in liquids. [1- 4] The results of Ref. [1] showed that the DTs in water are considerably, by a factor of ~1.5, higher than the corresponding values in air. The higher DT in water has been explained by scattering of the incident laser light by the vapor-liquid interface due to a vapor bubble formation in the subnanosecond timescale. However, these scattering processes obviously do not play any role for picosecond (ps) and femtosecond (fs) laser pulses since the pulses end before the liquid starts to be heated and vaporized.

In this work, we measured the DTs in water for gold and crystalline silicon. The targets were irradiated by fs (260 fs) and ps (7 ps) laser pulses of 1030 nm wavelength in single-shot and multi-shot regimes. The results are compared with data obtained in similar experiments in air. The influences of the pulse duration, surface reflectivity, focusing conditions and non-linear effects during laser pulse propagation in water are discussed.

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P5

Theoretical and experimental study of laser crystallization of thin amorphous layers of silicon under continuous laser irradiation

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The aim of our work is to crystallize 1–2 μm wide stripes of a-Si selectively etched from initial uniform layer of 400 nm thickness deposited on SiO_2. We have developed a numerical model that describes thermodynamic processes taking place during melting of material and solidification of crystals in continuous irradiation regime. Our results provide consistent description of the fabrication process that results in high quality, low-temperature Si waveguides. Final validation of a model represents an excellent starting point for simulation of ultrashort laser pulses and nonequilibrium melting that is a next stage of our research.

P6

Structuring of Kapton surface with ultrashort laser

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Nowadays there is huge demand for high precision processing of biocompatible polymers and, in particular, of polyimide for medical, electronics and industry applications. Pulsed laser ablation (PLA) is a well established method for surface processing and modification of polymers of different kinds. This process has extensively been investigated for UV region of electromagnetic spectra mainly with nanosecond laser pulses. However, polymer ablation in IR, Near- and Mid-IR spectral ranges is not widely been studied, especially in the regimes of ultrashort laser pulses when non-linear phenomena can play an important role in laser energy absorption. In this contribution, we report on a comparative study of surface structuring of polyimide (Kapton) by pico- and femtosecond laser pulses of 1030 nm wavelength. Accurate structuring of this biocompatible polymer is of importance for direct applications in prosthesis and other medical instruments. The ablation was performed with three lenses of different focal distances and the ablation thresholds have been determined. It was shown that femtosecond laser pulses are much more advantages for high-precision surface structuring compared to picosecond pulses.

P7

Reduction of Ag^+ ions by Polyamide 6 surface irradiated with ultrashort laser pulses for electroless copper deposition

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The process of laser-induced spatially-localised electroless copper deposition containing several steps is shown in Fig 1 a): laser excitation of the surface, wet chemical activation with a silver ionic solution, rinsing and electroless copper deposition in the bath. Ultrashort pulse laser was used for laser writing experiments, and a wide window of laser processing parameters was investigated to find out limitations of the technique and understand surface activations original. The substrate for metal deposition was polyamide 6 (PA6). Experimental results showed that the selective plating process is induced not by changes in surface morphology, but due to the change in the chemical composition after short pulse interaction with the polymer. X-ray Photo Electron (XPS) spectroscopy analysis was applied for mechanism investigation of selective copper deposition. XPS spectra were collected using the reference sample and samples after laser treatment and chemical activation with silver ionic solution steps.

XPS spectra showed variation in the atomic composition after the laser modification of PA6. In-depth spectral analysis of silver-related XPS-lines revealed that after laser treatment, PA6 gains reducing properties of metals ions (see Fig 1 b). Reduced silver (to metallic state) works as a catalyst for local electroless copper deposition. Part of the experiments was related to optimal laser and chemical parameters investigation. High processing speeds up to 4 m/s and plating pitch down to 25 µm has been achieved. [1].

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P8

Graphene Coating Generation by the Electrochemical and Femtosecond Laser-Assisted Reduction of Graphene Oxide

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Graphene oxide is a potential precursor for graphene. The mechanism of the electrochemical [1,2] as well as of a femtosecond laser-assisted reduction to reduced graphene oxide have been the focus of this present study. An electrochemical quartz crystal microbalance was utilized to perform mass-sensitive measurements during the electrochemical deposition of graphene oxide. The mechanism of the electrochemical reduction was investigated by in-situ attenuated total reflection FTIR spectroscopy. A multivariate method was applied for evaluating the spectra. It could be shown that the reduction occurs in two steps. At first graphene oxide is reduced to α-reduced graphene oxide. In a second step, it is reduced to β-reduced graphene oxide.

Moreover, the fs-laser-assisted reduction of graphene oxide coatings was demonstrated and investigated. The resulting reduced graphene oxide was characterized by Raman-spectroscopy and scanning electrochemical microscopy.

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P9

Graphene nano-strip generation by femtosecond laser-assisted apertureless scanning near-field optical lithography

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Graphene oxide is a promising precursor for the production of graphene. Photothermal and/or photochemical interactions using laser radiation may result in the photoreduction of graphene oxide [1]. In the present study, the reduction of graphene oxide is demonstrated by near-field femtosecond laser irradiation [2]. The laser beam was focused onto an atomic force microscope tip placed a few nanometers above the substrate. Near-field enhancement and/or thermal conduction below the tip induced the local reduction on graphene oxide nanosheets below the diffraction limit.

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P10

Hot electron electrochemistry induced by femtosecond laser pulses

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High intensity laser pulses can generate high densities of electrons in matter. One technologically important follow-up process is the deterministic multiphoton-electron coupling [1]. In the present work, the generation of high electron densities in a solid by high intensity femtosecond laser pulses [2-5] was investigated.

The subsequent emission of hot electrons into an electrolyte and the electrochemistry of intermediates was monitored as a function of laser and electrochemical parameters. Results may lead to a new understanding of the fundamentals of fast hot electron electrochemical kinetics, intermediate species electrochemistry [2-5], nanomedicine [6], materials machining in liquid contact [7], and the generation of colloidal solutions [8-10].

This study demonstrated that emitted hot electron charge densities are virtually independent of pulse duration for pulses below the electron-phonon coupling time (several picoseconds). However, it is strongly dependent on the incident laser fluence, and on the applied bias potential according to the Butler-Volmer and Tafel relationship of heterogeneous electron transfer kinetics.

With the observed picosecond duration current bursts of, nonequilibrium electrochemical experiments in the vicinity of a microelectrode can be performed allowing the investigation of e.g. intermediate species not accessible by conventional techniques, and the stabilization mechanisms of laser generated colloidal nanoparticles.

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