

7th European Conference on Applications of Femtosecond Lasers in Materials Science



FemtoMat 2017



**March 20 - 22, 2017
Mauterndorf Castle, Mauterndorf
Salzburg, Austria**

<http://www.esg-nano.ac.at/femtomat>

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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,

and the 6th FemtoMat 2015, Mauterndorf, Salzburg, Austria, March 2015, is the basis of the present

7th FemtoMat 2017, March 20 - 22, 2017, 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 2017

Wolfgang Kautek
(University of Vienna)



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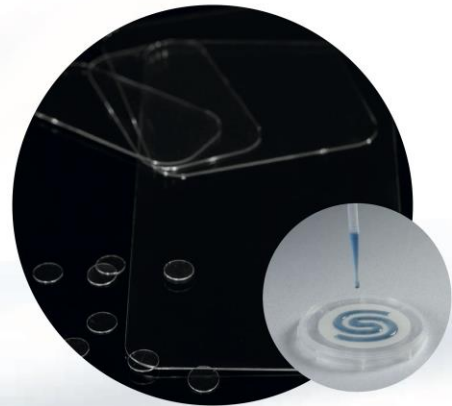
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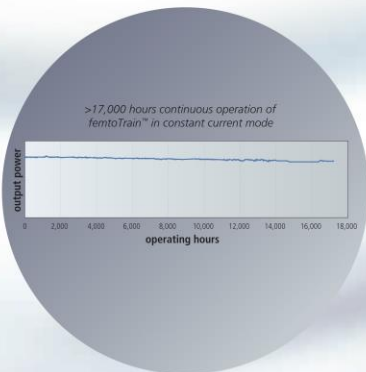
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Program

Monday, March 20, 2017

08:30 – 09:00 **Registration**

09:00 – 09:15 **Opening and greetings**

09:15 – 09:45 **L1** M. Sentis

Aix-Marseille University, F

"Femtosecond laser pulse energy deposition in fused silica"

09:45 – 10:15 **L2** R. Stoian

Université Jean Monnet, F

"Processing materials beyond diffraction limit using designer ultrafast laser radiation"

10:15 – 10:45 **L3** N.M. Bulgakova

HiLASE Centre, CZ

"Strength of laser-energy coupling to transparent dielectrics from ultrashort pulses with spatiotemporal features: Understanding and prediction through numerical modeling"

10:45 – 12:30 **Posters:** Lectures and session

12:30 – 16:30 **Free discussion**

16:30 – 16:45 **Coffee**

16:45 – 17:15 **L4** K. Sokolowski-Tinten

Universität Duisburg-Essen, D

"Ultrafast MeV electron diffraction studies of energy relaxation and transport in laser-excited thin film heterostructures"

17:15 – 17:45 **L5** H.P. Huber

Munich University of Applied Sciences, D

"Towards a better understanding of ultrafast laser processing of metals by experimental and simulated transient studies of reflectivity and absorption"

17:45 – 18:15 **L6** W. Perrie

University of Liverpool, UK

"Femtosecond inscription inside poly-methyl pentene with numerical aperture, wavelength and with polarised helical beams carrying orbital and spin angular momentum"

18:15 – 18:45 **L7** J. Krüger

Bundesanstalt für Materialforschung und -prüfung (BAM), D

"Femtosecond laser pulses for photovoltaic bottom-up strategies"

18:45 – 19:15 **L8** M. Pfeifenberger

Austrian Academy of Sciences, A

"The use of a femtosecond laser for rapid micro-mechanical sample preparation"

Tuesday, March 21, 2017

- 08:30 – 09:00 **L9** C. Spielmann
Friedrich Schiller University, D
"Interaction of nanostructured materials with intense mid-IR pulses"
- 09:00 – 09:30 **L10** M. Meunier
École Polytechnique de Montréal, CA
"Femtosecond interaction with plasmonic nanomaterials: Fundamentals and applications in nanomedicine"
- 09:30 – 10:00 **L11** A. Kanaev
University Paris 13, F
"Surface structuring of semiconductors in the regime of high-density electronic excitation"
- 10:00 – 10:30 **L12** B. Rethfeld
Technische Universität Kaiserslautern, D
"Relaxation cascade of laser-excited electrons in solids"
- 10:30 – 10:45 **Coffee**
- 10:45 – 11:15 **L13** P. Balling
Aarhus University, DK
"Ultrashort-pulse laser excitation of materials: Fundamentals and applications"
- 11:15 – 11:45 **L14** L.V. Zhigilei
University of Virginia, USA
"Computational study of the generation of nanoparticles and surface nanocrystallization by short pulse laser irradiation of metal targets in liquid environment"
- 11:45 – 12:15 **L15** A. Fuerbach
Macquarie University, AUS
"Femtosecond laser direct-writing of integrated photonic devices"
- 12:15 – 16:30 **Free discussion**
- 16:30 – 16:45 **Coffee**
- 16:45 – 17:15 **L16** W. Rudolph
University of New Mexico, USA
"Nonlinear optical elements based on aperiodic dielectric layers - frequency tripling mirror (FTM)"
- 17:15 – 17:45 **L17** T. Winkler
University of Kassel, D
"Laser amplification in laser excited dielectric materials"
- 17:45 – 18:15 **L18** E.L. Gurevich
Ruhr-Universität Bochum, D
"Combined hydrodynamic-plasmonic model of the laser induced periodic surface structures (LIPSS) formation"
- 18:15 – 18:45 **L19** T. Itina
University of Lyon, F
"Ultra-short laser structuring of glasses: Predictive modeling insights"
- 18:45 – 19:15 **L20** P. Dombi
Wigner Research Centre for Physics, HU
"Measurement of nanoplasmonic field enhancement with ultrafast photoemission"

Wednesday, March 22, 2017

- 08:30 – 09:00 **L21** C. Grigoropoulos
University of California, Berkeley, USA
"Ultrafast Laser Two- and Three-Dimensional Nano Patterning"
- 09:00 – 09:30 **L22** G. O'Connor
National University of Ireland Galway, IRL
"Nanostructure-enabled precision structuring using ultrashort laser processes"
- 09:30 – 10:00 **L23** P. Simon
Laser-Laboratorium Göttingen e.V., D
"Nano-structure formation on gold and silicon surfaces by laser irradiation"
- 10:00 – 10:30 **L24** A. Bulgakov
S.S. Kutateladze Institute of Thermophysics SB RAS, RUS
"Acceleration of ions emitted from ultrashort-laser-irradiated surfaces in the photoelectron-induced electric field"
- 10:30 – 10:45 **Coffee**
- 10:45 – 11:15 **L25** M. Böhmler
neaspac GmbH, D
"Ultra-fast nano-FTIR spectroscopy: a new time-resolved view to the nanoworld"
- 11:15 – 11:45 **L26** C. Plamadeala
Johannes Kepler University Linz, A
"Bio-inspired microstructures for directional transport of liquids fabricated by two-photon polymerization"
- 11:45 – 12:15 **L27** R. Haglund
Vanderbilt University, USA
"A nanostructured metadvice based on phase-changing oxide"
- 12:15 – 12:30 **Closing remarks**

Lecture Abstracts

L1

Femtosecond laser pulse energy deposition in fused silica (Invited)

M. Sentis, O. Utéza, M. Lebugle, N. Varkentina, N. Sanner

Aix Marseille University, CNRS, LP3, UMR7341, FR

A femtosecond laser is a powerful fabrication tool able to deposit energy for local transformation of a material with high spatial resolution and precision [1]. In this context, the knowledge of the details of the laser energy deposition in the material is of prime importance because the induced effects finally leading to the macroscopic material changes closely depend on its characteristics.

In this work, we use near-infrared femtosecond pulses to drive highly-localized absorption in wide band-gap transparent dielectrics [1, 2]. Briefly, the pulse leading edge is absorbed by non-linear photo-ionization processes, providing seed free electrons in the conduction band that are further accelerated through inverse Bremsstrahlung and multiplied by impact ionization. A plasma of high electron density is thus created in the first layers of the surface that absorbs the trailing part of the pulse [2].

Our purpose is to measure with high precision the evolution of the transient optical properties of the laser-generated electron-hole plasma in order to uncover the details of energy deposition on dielectrics. We will present and discuss recent pump-probe measurements performed at the surface of dielectrics and able to track i) the quantity of laser energy E_{abs} that is really deposited in the material [3] and ii) the dynamics of this phenomenon, i.e. the time-resolved history of energy deposition process [4].

Finally, the full diagnostic of energy balance at the surface of the dielectrics allow us to define some useful guidelines for the laser tool user in charge of the development of efficient processes of micromachining of dielectric materials [5].

References:

- [1] R. Gattass, E. Mazur, Femtosecond laser micromachining in transparent materials, *Nat. Phot.* 2, 219 (2008).
- [2] E. Gamaly, E., Femtosecond laser-matter interactions. Pan Stanford Publishing, (2011).
- [3] N. Varkentina, N. Sanner, M. Lebugle, M. Sentis, O. Utéza, Absorption of a single 500fs laser pulse at the surface of fused silica: Energy balance and ablation efficiency, *J. Appl. Phys.* 114, 173105, (2013).
- [4] M. Lebugle, N. Sanner, N. Varkentina, M. Sentis, O. Utéza, Dynamics of femtosecond laser absorption of fused silica in the ablation regime, *J. Appl. Phys.* 116, 063105, (2014).
- [5] M. Lebugle, N. Sanner, O. Utéza, M. Sentis, Guidelines for efficient direct ablation of dielectrics with single femtosecond pulses - *Appl. Phys. A* 114 (1), 129-142, (2014).

Processing materials beyond diffraction limit using designer ultrafast laser radiation (Invited)

R. Stoian

Université de Lyon, Université Jean Monnet, Laboratoire Hubert Curien, FR

Bypassing diffraction limit in laser material structuring is a key issue for a new range of applications in optics and mechanics requiring optical access to the nanoscale. Enabled by the nonlinearity of interaction, ultrafast lasers show remarkable effectiveness in localizing light on subwavelength scales, building up in many cases on a collective carrier response on surfaces and in the bulk. The 3D capability has a particular interest as ultrafast laser interaction with transparent materials can achieve optical functions by space-design of embedded structural transformations with changes of the dielectric function. Control of laser interaction by beam design can drive selected physical paths and geometries, and we will focus here on structural evolutions and dimensional scales enabled by spatiotemporal beam shaping. Essential for refractive index engineering, laser-induced matter transformation can be significantly influenced by the level of the energy deposition, critically dependent on the pulse temporal envelope. Photoionization can be regulated, leading to unprecedented localization of laser energy. Pulse temporal and spatial design can achieve index structures on scales approaching 100 nm, either in direct focusing or self-organization schemes in model fused silica. We follow specific dynamics of electronic relaxation in confinement conditions and point out characteristic times of energy deposition, serving as guidelines for control. Fast electronically-induced structural changes or slow thermodynamic transformations can be discriminated. Concepts of non-diffractive beam excitation can additionally take advantage of this localization and achieve unprecedented high aspect ratio structuring. From the application point of view, the mid-infrared spectral range carries a strong potential in sensing and imaging. Extrapolation of controlled laser-induced structural modification towards mid-infrared materials can achieve strong index contrast on micron and submicron scales. We demonstrated linear and nonlinear 3D optical functions in chalcogenide glasses where light transport in mid-infrared can be efficiently achieved with large area modes, and the field distributions can be non-perturbatively accessed.

L3

Strength of laser-energy coupling to transparent dielectrics from ultrashort pulses with spatiotemporal features: Understanding and prediction through numerical modeling (Invited)

N.M. Bulgakova^{1,2}, V.P. Zhukov^{3,4}, A.M. Rubenchik⁵, Y. Morova⁶, S. Aktürk⁶

¹ HiLASE Centre, Institute of Physics ASCR, CZ

² S.S. Kutateladze Institute of Thermophysics SB RAS, RU

³ Institute of Computational Technologies SB RAS, RU

⁴ Novosibirsk State Technical University, RU

⁵ Lawrence Livermore National Laboratory, CA, USA

⁶ Istanbul Technical University, Department of Physics, TR

Developing techniques for manipulation and control over spatiotemporal shape of laser beams opens new horizons for more precise and cleaner processing of various materials. These techniques are of special interest for volumetric processing of optical glasses and crystals with inducing structural changes at micro- and nanoscales as demanded in a variety of applications.

In this talk we will focus on sophisticated modeling, based on Maxwell's equations, of propagation of ultrashort laser beams with spatiotemporal features in transparent solids to get insight into volumetric laser energy coupling that can induce permanent modification in a localized zone inside material bulk. The model predictions will be demonstrated for radially-polarized doughnut-shaped pulses and for Gaussian pulses with tilted front and lighthouse effect. It will be shown that doughnut-shaped laser pulses enable much more efficient energy coupling into a highly localized near-focal zone as compared to Gaussian beams of the same energy. Furthermore, at certain irradiation conditions, they can provide an implosion process that can bring matter to extreme thermodynamic states culminating in exotic structural changes. Some peculiarities of multipulse irradiation regimes will also be discussed.

Ultrafast MeV electron diffraction studies of energy relaxation and transport in laser-excited thin film heterostructures

(Invited)

K. Sokolowski-Tinten¹, R. Li², A.H. Reid², S.P. Weathersby², F. Quirin¹, T. Chase², Z. Shen², R. Coffee², J. Corbett², N. Hartmann², C. Hast², R. Hettel², M. Horn von Hoegen¹, D. Janoschka¹, M. Jermann¹, J.R. Lewandowski², M. Ligges¹, F. Meyer zu Heringdorf¹, M. Mo², X. Shen², T. Vecchione², C. Witt¹, J. Wu², Q. Zheng², H. Dürr², X. Wang²

¹ University of Duisburg-Essen, Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), D

² SLAC National Accelerator Laboratory, CA, USA

Electron-phonon and phonon-phonon interactions determine electronic and thermal transport properties of materials and are responsible for the transfer and equilibration of electronic excess energy to/with the lattice. With decreasing device dimensions an understanding of these processes in nanoscale materials is becoming increasingly important. Here we report the results of time-resolved MeV transmission electron diffraction experiments carried out at the recently established femtosecond UED-facility at SLAC Nat. Lab, addressing the incoherent structural response of different thin-film heterostructures after femtosecond laser excitation.

L5

Towards a better understanding of ultrafast laser processing of metals by experimental and simulated transient studies of reflectivity and absorption

H.P. Huber

Munich University of Applied Sciences, D

Ultrashort pulsed lasers offer a high potential for precise and efficient material processing. A deep and thorough understanding of the fundamental laser-material interaction aspects is of great importance to optimize laser processes for industry. The transient pulse reflectivity in conjunction with the transient absorption decisively influences the laser-material interaction. Direct measurements of the absorption properties, however, e.g. by ultrafast time-resolved ellipsometry are missing to date.

We present a unique pump-probe ellipsometry microscope allowing the determination of the transient complex refractive index $N = n - ik$ of laser irradiated materials with a sub ps temporal resolution and an accuracy <3%. Measurements were performed on industrial relevant metals (Cu, Al, stainless steel AISI 304) at laser fluences near the ablation threshold. The measured refractive index n and extinction coefficient k on copper are supported by a simulation including the accurate description of optical, thermal (two-temperature model) and thermomechanical properties. A comparison between measured time-resolved optical properties and results of the simulation reveals underlying physical mechanisms in the first tens of picoseconds. Around delay time zero thermally excited d-band electrons create a steep increase in n and k . Up to about 10 ps, the material expansion decreases density and the absorption k . Additionally, the electron-ion thermalisation time can be attributed to a minimum of k at ~10 ps. After 10 ps the slowly rising k indicates the surface cooling-down process.

In conclusion, the combination of pump-probe ellipsometric experiments and finite element simulations gives new and deep insights in the absorption change of the pump pulse or subsequent pulses irradiating the sample on ultrashort timescales. These insights pave the road towards a better understanding of the pulse duration dependent laser ablation efficiency, double or burst mode laser ablation and lattice modifications in the first ps after the laser pulse impact.

Femtosecond inscription inside poly-methyl pentene with numerical aperture, wavelength and with polarised helical beams carrying orbital and spin angular momentum

G. Zhu¹, B. Dorin², W. Perrie¹, P. Scully², P. Parkinson², D. Liu³, S. Edwardson¹, G. Dearden¹

¹ University of Liverpool, UK

² University of Manchester, Photon Science Institute, UK

³ Hubei University of Science and Technology, CN

This paper reports on a study of femtosecond laser inscription in Polymethyl Pentene in the NUV at 387.5 nm, at visible wavelengths 550 - 700 nm, with NA in the range 0.1 - 0.7. Low NA inscription at all wavelengths resulted in a correspondingly low refractive index modulation, $\Delta n = 1 \times 10^{-4}$ whereas NA = 0.4 inscription resulted in an order of magnitude increase in refractive index modulation, $\Delta n = 2.3 \times 10^{-3}$. Volume Bragg Gratings inscribed at 387.5 nm with 200 lines/mm showed a first order diffraction efficiency > 68.5%, the highest yet reported in this polymer. The critical power for self-focusing in PMP at 387.5 nm, estimated from the threshold for supercontinuum generation was found to be 2.2 MW inferring that the non-linear refractive index is $n_2 = 2 \times 10^{-16} \text{ cm}^2 \text{ W}^{-1}$. A spatial light modulator addressed with spiral Computer Generated Holograms created helical wavefronts carrying Orbital Angular Momentum (OAM). Inscription using NUV spiral phase beams carrying OAM at 387.5 nm with linear and circular polarizations was investigated for the first time in PMP. Critical powers for SF with spiral beams scale linearly with topological charge m and linearly polarized spiral beams always couple more strongly than circular due to polarization dependent self-focusing. With high states of OAM, complex filamentary structures converging to the focus are observed which collapse to multiple filaments. Rotation of these filaments is detected near the focal plane due to the twisting wavefronts.

L7

Femtosecond laser pulses for photovoltaic bottom-up strategies (Invited)

J. Krüger

Bundesanstalt für Materialforschung und -prüfung (BAM), D

A bottom-up approach is presented for the production of arrays of indium islands on a molybdenum layer on glass using 30-fs laser pulses at 790 nm wavelength [1]. The indium islands can serve as micro-sized precursors for indium compounds such as copper-indium-gallium-diselenide (CIGSe) used in photovoltaics. Molybdenum is the standard back contact material of CIGSe solar cells. Femtosecond laser ablation of glass and a subsequent deposition of a molybdenum film or direct laser processing of the molybdenum film both allow the preferential nucleation and growth of indium islands at the predefined locations in a following indium-based physical vapor deposition (PVD) process. A proper choice of laser and deposition parameters ensures the controlled growth of indium islands exclusively at the laser ablated spots. Based on a statistical analysis, these results are compared to the non-structured molybdenum surface, leading to randomly grown indium islands after PVD.

References:

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The use of a femtosecond laser for rapid micro-mechanical sample preparation

M.J. Pfeifenberger

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Focused ion beam (FIB) machining represents the state-of-the-art fabrication technique for the preparation of miniaturized mechanical samples, due to its high precision and availability. Nevertheless, it poses also a bottleneck on account of the small removal rates. Further, ion implantation leads to damage of the near surface material. Employing a femtosecond laser for material ablation offers removal rates multiple orders of magnitude higher compared to a FIB and it exerts little to no thermal influence on the surrounding material. Therefore, a combination of the two techniques offers an ideal tool for micrometer-sized sample preparation. A prototype is developed on the basis of the Zeiss Auriga Laser platform, where the laser structuring is conducted in a separated airlock chamber to prevent contamination of the main chamber and to allow different atmospheric conditions. The performance of the novel tool is demonstrated with two model systems.

First, cantilevers with a length of several hundred micrometers have been machined into rolled tungsten foils employing both a nanosecond and a femtosecond laser. The quality of the resulting samples in terms of feature size and surface roughness as well as the influence on the microstructure is analyzed and compared. Second, tensile specimens from spruce wood are fabricated using a femtosecond laser and an ion slicing device. A critical comparison of the results exhibits in both applications a clear advantage of the femtosecond laser technique.

Interaction of nanostructured materials with intense mid-IR pulses (Invited)

R. Hollinger^{1,2}, Z. Samosonva^{1,2}, S. Höfer¹, D. Kartashov¹, S. Alisauskas³, V. Shumakova³,
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² Helmholtz Institute Jena, D

³ TU Vienna, Photonics Institute, A

⁴ FSU Jena, Institute of Solid State Physics, D

⁵ Technical University Braunschweig & PTB, Braunschweig, LENA Joint Laboratory, D

Nonlinear optics with few-cycle laser pulses in nanoscale solids is of great importance for fundamental research and practical applications. Nanostructures as targets enhance not only the optical frequency conversion efficiency, but they are also the key element for the efficient generation of ultrafast X-ray pulses from relativistic plasmas. The enhanced nonlinear response in the different regimes is a consequence of e.g. local field enhancement or more favorable propagation conditions in a plasma generated from nanostructured materials. Additionally the increased surface area-to-volume ratio leads to a drastic enhancement of absorption and emission and enables rapid generation of highly overcritical plasma densities due to volumetric heating. For all these experiments a precise determination of the damage threshold is crucial, because it not only defines the maximum intensity for nondestructive experiments, but also the final plasma parameters in destructive experiments.

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 a lowering of the damage threshold. However, due to long wavelength tunneling excitation can become the dominant absorption mechanism. So far, the nonlinear absorption has been extensively experimentally studied for VIS/near-IR driver pulses, but almost no studies exist for pulses in the few μm range.

Recently, we started to explore the nonlinear interactions of intense femtosecond laser pulses with single nanowires and arrays of nanowires made of various materials including ZnO. Using frequency-tunable ultrafast laser sources developed at TU Wien and FSU Jena, we studied the wavelength dependent nonlinear absorption of nanostructured and bulk samples in a wide intensity range. E.g. by tuning the wavelength of the femtosecond pulses from the visible to the mid-IR, we have explored resonant and off-resonant multiphoton regimes, with a particular attention on the possibility of nanowire excitation in the case of tunneling ionization. As a further proof of the different absorption mechanism for different wavelengths and morphologies of the structures we will present x-ray spectra which are sensitive to the absorption mechanism. First results of these studies will be presented at the conference.

**Femtosecond interaction with plasmonic nanomaterials:
Fundamentals and applications in nanomedicine**
(Invited)

M. Meunier

École Polytechnique de Montréal, CAN

Irradiating metallic nanostructures with a femtosecond laser beam produces highly localized processes on the nanoscale in the surrounding medium. This particular process is mainly attributed to the surface plasmon resonance of the nanostructures. When these nanomaterials are imbedded in a biological media, their irradiation by a femtosecond laser could results in a highly localized plasma, heat production and mechanical effects yielding to the nanosurgery of cells. Recent developments of the fundamentals and applications of nanoplasmonics enhanced ultrafast laser nanosurgery of living cells will be presented. Modeling and experiments were developed to understand the basic phenomena occurring during ultrafast laser irradiation of plasmonics nanostructures. We will describe an in-silico rational design approach based on theoretical modeling that optimizes cavitation without breaking the nanostructure. Applications of laser optoporation and transfection of cells in various field of nanomedicine will be discussed. This technique has been successfully used to locally stimulate neurons.

References:

- 1 J. Baumgart et al Biomaterials, 33, 2345-2350 (2012).
- 2 E. Boulais et al Nanoletters, 12, 4763-4769 (2012).
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- 9 F. Lavoie-Cardinal et al Scientific Reports, 6, 20619 (2016).
- 10 R Lachaine et al Nano letters 16 (5), 3187-3194 (2016)
- 11 R. Lachaine et al ACS Photonics, accepted (2016).

L11

Surface structuring of semiconductors in the regime of high-density electronic excitation (Invited)

L. Museur¹, A. Manousaki³, D. Anglos³, G. Tsididis³, A. Kanaev²

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FR

³ Institute of Electronic Structure & Laser, Foundation for Research and Technology - Hellas (IESL-FORTH), GR

The talk is devoted to the surface structuring of semiconductors in the regime of high density electronic excitation above Mott transition. The irradiation of ZnO and TiO₂ monocrystals and thin films was performed with femtosecond KrF laser, which photons energy is above that of the band gap. The nanostructuring sensitively depends on the crystalline planes orientation, laser polarization, fluence and dose. Ripples, 10-nm nanoholes interconnected with straight and zig-zag lines and 100 nm nanocraters were observed depending on the irradiation conditions. The mechanism of the structuring is discussed.

Relaxation cascade of laser-excited electrons in solids (Invited)

B. Rethfeld, S. Weber, A. Ramer, N. Brouwer

Technical University of Kaiserslautern, Department of Physics and OPTIMAS research center, D

During femtosecond laser irradiation of solids, mainly the electrons in the material gain energy. Free electrons in the conduction band of a metal can directly absorb photons. In semiconductors and dielectrics, on the other hand, a band gap has to be overcome first, before further energy gain from the laser beam is possible. These absorption processes transfer the electronic system to a state of strong thermodynamic nonequilibrium.

A sequence of different relaxation processes transfers the material into a new equilibrium. They occur on different timescales and partially influence each other. Usually, intraband thermalization is the fastest of these processes. Here, electrons in each excited band establish a new Fermi distribution of elevated temperature. In case of several excited electron bands, these temperatures equilibrate due to interband energy exchange. Additionally, particles have to be exchanged between the bands in order to establish a joint chemical potential of all electrons.

Finally, ultrafast electron-phonon collisions lead to the temperature relaxation of hot electrons and the initially cold lattice. The energy transfer due to these collisions is completed on rather long picosecond timescales, while the collisions themselves occur on a femtosecond timescale. These collisions, therefore, also influence the relaxation processes within the electronic system.

We show examples of the various relaxation processes, discuss their timescales and present examples of their mutual influence.

L13

Ultrashort-pulse laser excitation of materials: Fundamentals and applications (Invited)

P. Balling

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Ultrashort laser pulses open up for new material interaction pathways. In this presentation, recent investigations of short-pulse excited dielectric materials are presented. Experimentally, time-resolved optical experiments (reflectivity and spectral interferometry) measure the dynamics of the excitation, and the observations are compared to theoretical models based on the so-called extended multiple-rate-equation model. In addition, the applications of short-pulse laser excitation for the mapping of optical near fields will be discussed.

Computational study of the generation of nanoparticles and surface nanocrystallization by short pulse laser irradiation of metal targets in liquid environment

(Invited)

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Laser ablation in liquid environment is actively used for generation of clean colloidal nanoparticles with unique shapes and functionalities suitable for applications in various fields, including biomedicine, chemical catalysis, and plasmonics. Moreover, the interaction of the hot transiently melted surface of the irradiated target with liquid environment can contribute to the rapid quenching of the surface structures, thus creating conditions for stronger undercooling and formation of highly nonequilibrium surface microstructure. In this presentation, we report the results of atomistic simulations of laser interactions with metal targets in liquid environment aimed at revealing the mechanisms of nanoparticle formation in pulsed laser ablation in liquids and evaluation of the conditions leading to surface nanocrystallization.

Two distinct mechanisms of the nanoparticle formation are predicted in the simulations of laser ablation in liquids: (1) the nucleation and growth of small (mostly ≤ 10 nm) nanoparticles in the metal-water mixing region and (2) the formation of larger (tens of nm) nanoparticles through the breakup of the superheated molten metal layer triggered by the emergence of complex morphological features attributed to the Rayleigh-Taylor instability of the interface between the superheated metal layer and the supercritical water. The computational prediction of the two mechanisms of nanoparticle formation yielding nanoparticles with different characteristic sizes is consistent with experimental observations of two distinct nanoparticle populations appearing at different stages of the ablation process.

The effect of the liquid environment on the laser-induced surface nanocrystallization is investigated in the regime of melting and resolidification, below the thresholds for laser spallation and ablation. The presence of a liquid overlayer is found to provide an additional pathway for cooling through the heat conduction to the overlayer and facilitate the formation of nanocrystalline structure in a region of the metal target adjacent to the overlayer. The nanocrystalline layers generated by laser processing of single-crystal metal targets in liquid environment are characterized by a high density of stacking faults, twin boundaries, and point defects produced in the course of the rapid resolidification.

L15

Femtosecond laser direct-writing of integrated photonic devices (Invited)

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M. Withford

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Tightly focused femtosecond laser pulses can be used to alter the refractive index of virtually all optical glasses. As the laser-induced modification is spatially limited to the focal volume of the writing beam, this technique enables the fabrication of fully three-dimensional photonic structures and devices that are automatically embedded within the host material. While it is well understood that the laser-material interaction process is initiated by nonlinear, typically multiphoton absorption, the actual mechanism that results in an increase or sometimes decrease of the refractive index of the glass strongly depends on the composition of the material and the process parameters and is still subject to scientific studies.

In this talk, we present an overview of our recent work aimed at uncovering the physical and chemical processes that contribute to the observed material modification. Raman microscopy and electron microprobe analysis was used to study the induced modifications that occur within the glass matrix and the influence of atomic species migration forced by the femtosecond laser writing beam. We demonstrate that a detailed understanding of the physical processes that underlie the femtosecond laser direct-write technique enables the fabrication of three-dimensional photonic devices with unprecedented performance.

**Nonlinear optical elements based on aperiodic dielectric layers -
frequency tripling mirror (FTM)**
(Invited)

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A frequency tripling mirror for femtosecond laser pulses based on aperiodic stacks of dielectric layers is demonstrated. We describe scaling of conversion efficiency and bandwidth with respect to the number of layers and layer properties. Material science problems are discussed whose solution would make these novel mirrors competitive with crystal optics.

L17

Laser amplification in laser excited dielectric materials (Invited)

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Laser excitation of transparent dielectrics like sapphire or fused silica by ultra-short laser pulses usually cause a transition of the material to a transient metallic-like state. This is caused by the creation of a high carrier density in the conduction band, which is the first step in laser-material processing of dielectrics. This metallic state is typically identified by a high absorption and reflectivity as shown in a manifold of experiments.

In our recent studies, which are based on an in-line pump probe and a common-path spectral interference setup, we show that under appropriate conditions, the strong absorption in a thin laser excited dielectric sample is replaced by optical gain. The optical gain is coherent, directed and based on stimulated three-photon emission. We present energetic, temporal and spectral dependencies and properties of the laser excitation in a thin excited sapphire sample.

Combined hydrodynamic-plasmonic model of the laser induced periodic surface structures (LIPSS) formation

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Formation mechanisms of the Laser Induced Periodic Surface Structures (LIPSS) are discussed since the pattern was discovered and first published in 1965. The suggested models of the LIPSS formation can be separated in two classes: plasmonic, involving interaction between the surface plasmons and the incident laser light, and hydrodynamic, which are based on the analysis of hydrodynamic instabilities in the thin molten layer on the surface. These both approaches can explain some of the experimentally-observed facts but fail to explain the whole process. Here we suggest a coupled approach, in which the initial perturbation for the instability is selected by the weak plasmons and amplified by hydrodynamic mechanisms.

L19

Ultra-short laser structuring of glasses: Predictive modeling insights (Invited)

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Short and ultrashort laser pulses are particularly attractive for micro- and nano-structuring of glasses. Among the most promising applications, we can mention the formation of periodic and non-periodic 2D and 3D nanostructures that can be used not only in optics and photonics (optical memories, optical components, metamaterials, photonics crystals), but also in security, nanofluidics, medicine, etc.

Numerous experiments were focused on the investigation of the main features of ultra-short laser processing of dielectric materials [1-2]. It was demonstrated that a particular parameter window is often preferable to obtain the desired results and that different types of permanent material modification could be induced in glasses depending on the laser irradiation conditions. In these experiments, both single pulse and multi-pulse laser irradiation was used. However, many involved processes are still under discussion. To better control over the laser-based processing, a comprehensive and predictive modeling is developed and the results will be presented in this talk. In our modeling, first light propagation is modeled in a consistent way with possible non-linear material ionization due to the local field enhancement. Then; we consider the main photo-thermal processes taking place during ultra-short laser irradiation and thermo-mechanical wave propagation. As a result, spatiotemporal evolutions of temperature, pressure and density are calculated, which accompany the refractive index change over a broad range of timescales from femtoseconds to few microseconds as a function of laser parameters such as pulse energy, pulse duration, laser wavelength, numerical aperture, number of pulses and repetition rate. By applying this model, we study the role of the laser experimental conditions and the one of material properties. The physical processes such as thermo-mechanical effects and heat accumulation are also investigated and the parameter windows required for efficient laser treatment are explained.

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Measurement of nanoplasmonic field enhancement with ultrafast photoemission
(Invited)

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Probing nanooptical near-fields is a major challenge in plasmonics. Here, we demonstrate an experimental method utilizing ultrafast photoemission from plasmonic nanostructures that is capable of probing the maximum nanoplasmonic field enhancement in any metallic surface environment. Directly measured field enhancement values for various samples are in good agreement with detailed finite-difference time-domain simulations. These results establish ultrafast plasmonic photoelectrons as versatile probes for nanoplasmonic near-fields.

L21

Ultrafast laser two- and three-dimensional nano patterning (Invited)

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This talk aims at reviewing work conducted at the Laser Thermal Laboratory on the nanoscale laser modification of materials using ultrafast laser pulses. We applied 2-photon laser ablation to write sub-diffraction nanoscale chemical patterns into ultrathin polymer films under ambient conditions. Poly(ethylene glycol) methacrylate brush layers were prepared on quartz substrates via surface initiated atom transfer radical polymerization (SI-ATRP) and ablated to expose the underlying substrate using the non-linear 2-photon absorbance of a femtosecond laser beam. Ablated features were chemically distinct and amenable to chemical modification.

Although adhesive interactions between cells and nanostructured interfaces have been studied extensively, there is a paucity of data on how similar interfaces repel cells, and direct migration and cell organization. We carried out a quantitative study addressing cell response to nanoscale craters patterned with various aspect ratios and differential spacing (i.e., pitch) fabricated via multiphoton ablation lithography. The nanostructured surfaces altered focal adhesion distribution affecting cell adhesion, morphology, and migration.

We have devised a new method for fabricating high aspect ratio patterns of varying height by using two-photon polymerization process in order to study contact guidance and directed growth of biological cells. Cell morphology on fiber scaffolds is influenced by the pattern of actin microfilament bundles. Cells experienced different strength of contact guidance depending on the ridge height. A significant effect on cell alignment, directionality of migration as well as on cell morphology and motility was observed. Cell contractility was examined microscopically in order to measure contractile forces generated by individual cells on self-standing fiber scaffolds.

Finally, we utilized ultrafast laser beam processing for generating scaffold multi-scale structures with 100 nm feature resolution. Once the template is constructed, directed self-assembly of block copolymers is used to produce three-dimensional materials with tailored functionality where pattern amplification in order to push the length scale to the 10 nm regime. The directed self-assembly of block copolymers is a parallel process and, as such, particularly over the fundamental length scales of concern in these studies, is quite rapid.

Nanostructure-enabled precision structuring using ultrashort laser processes (Invited)

G. O'Connor

National University of Ireland Galway, NCLA Laser Group/CURAM, IRL

Repetitive short and ultrashort laser sources are important reconfigurable tools for precision structuring of materials using future high throughput manufacturing platforms. The potential for precision laser processes depends on the applied fluence. Non-thermal multipulse processes are described. The application of the first low fluence, ultrashort, laser pulse is known to lead to the formation of a nanostructure. The interaction of the second and subsequent pulses with this nanostructure leads to precision material re-structuring and/or ablation. The application of such nanostructure-enabled multipulse laser processes is demonstrated in laser induced periodic surface structures, selective patterning and structuring of ITO, and surface structuring of the molybdenum aluminium bi-layer system. We identify the initial nanostructures to act as possible plasmonic lenses in the onset of laser induced periodic surface structures. In precision selective patterning of ITO, the initial nanostructures lead to non-thermal, precision, photomechanical ablation of thin films using cyclic compressive stresses. Laser-thin film-nanostructure interactions lead to submicron nanoscale structuring of glass and aluminium. Real time monitoring of laser surface interactions contribute to identifying the possible mechanisms by which nanostructure positively impacts the precision of laser processing.

L23

Nano-structure formation on gold and silicon surfaces by laser irradiation (Invited)

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Nano-structured surfaces can give rise to new material properties with very special electrical, mechanical, optical or biological characteristics. Depending on the specific material parameters and the morphology of the structures, special functionalities can be obtained offering the potential for numerous novel applications. These include the formation of super-hydrophobic behavior, particular tribological or cell growth properties and decorative or security features for the refinement of precious goods.

A full understanding of the fundamental microscopic mechanisms underlying the generation of nanostructures on surfaces is however still missing. On the other hand, this knowledge is extremely valuable to be able to fabricate tailored surface structures with sub-100 nm resolution, thus opening up new possibilities for the creation of particular functionalities. Therefore, our objective is to study the dynamics of the formation of periodic nano-structures.

Detailed theoretical simulation of the process was carried out by a „first principles two temperature model molecular dynamics“ (FP-TTM-MD) approach including an ab-initio calculation of the field induced modification of the potential surfaces and the electron dynamics. Moreover, the implementation of a dynamic reflectivity of the irradiated target surface will be discussed.

A carefully designed experimental strategy makes it possible to compare theory and experiment on the same temporal and spatial scale. We obtain excellent agreement between the simulation and the experimental results.

Acceleration of ions emitted from ultrashort-laser-irradiated surfaces in the photoelectron-induced electric field

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We present a model of acceleration of ions emitted from surfaces upon femtosecond laser irradiation at relatively low fluences, near or even below the ablation thresholds. The model assumes that the ions are accelerated in an electric field which is generated near the irradiated surface for a short time (~ 100 ps) due to laser-induced electron emission. The acceleration mechanism has a general character and does not depend on the particle emission mechanism. Using a drift-diffusion approach, we have performed modeling of silicon charging under 800-nm, 100-fs laser irradiation and thus estimated the generated fields, the characteristic times of their existence, and the maximal achievable ion velocities. The model predicts a momentum scaling law for ions of different masses with their velocities dependent on the irradiated spot size. To verify the model, we have performed a mass spectrometric study of clusters emitted from fs-laser-irradiated silicon. The maximum cluster velocities determined from the time-of-flight distributions are found to clearly scale with momentum and agree well with the model predictions. We thus demonstrate that the momentum scaling law is a general feature for fs-laser-produced ions, regardless of the mechanism of their emission.

L25

Ultra-fast nano-FTIR spectroscopy: a new time-resolved view to the nanoworld

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For the first time, ultra-fast nano-FTIR spectroscopy enables the investigation of materials in the infrared regime with spatial and temporal resolution beyond the classical limits. The new technology combines the unmatched spatial resolution of scattering-type scanning near-field optical microscopy with the high time-resolution of the pump-probe method. It relies on the unique properties of a DFG light source based on femtosecond lasers. This promising combination already demonstrated its power in the fields of 2D materials and semiconductor nano-devices.

A detailed insight of ultra-fast nano-FTIR spectroscopy will be provided and the first application examples will be reported.

Bio-inspired microstructures for directional transport of liquids fabricated by two-photon polymerization

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Nature always served as inspiration for scientists, helping them to solve a large diversity of technical problems. In our case, we are interested in the directional transport of liquids and as a role-model for this application we used flat bugs. We present arrays of microstructures that mimic the micro-ornamentation from the bugs' cuticle. These polymer microstructures are written by a Ti-sapphire femtosecond-laser which is focused into a liquid acrylate-based photopolymer containing a photoinitiator. A good directionality of liquid transport was achieved, directly controlled by the direction of the pointed microstructures at the surface. These results could therefore be interesting for applications in microfluidics and tribology.

L27

A nanostructured metadvice based on phase-changing oxide (Invited)

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There is tremendous interest at present in making use of phase-changing materials (PCMs) as the active components that enable the fabrication of plasmonic metamaterials on a practical scale.[1] The challenge is especially acute in the visible and near-infrared, spectral regions where finding appropriate PCMs and integrating them into chip-scale structures given the requisite size and fabrication dimensions requires exquisite control of materials processing and electron-beam lithography (EBL). Vanadium dioxide (VO₂) has attracted particular interest as the PCM for switchable metamaterials because of its reversible insulator-to-metal transition that can be triggered thermally, optically or electrically.

In this presentation, we first describe efforts undertaken to meet these challenges by building simple hetero-dimer modulators involving gold and VO₂ nanodisks, using a two-step EBL process and pulsed laser deposition to produce stoichiometric, switching VO₂. We were then able to measure the interaction strength between the gold and VO₂ nanodisks by monitoring the shift in the surface-plasmon resonance as a function of the edge-to-edge distance between them, down to 14 nm.[2] In the experiment currently underway, we fabricated an array of overall dimensions roughly 24x24 μm² (7200 pixels) in which VO₂ nanocylinders (120x120x37 nm³) in a nearly square arrangement were prepared by EBL, physical vapor deposition (RF magnetron sputtering) and a fast annealing step. In a second EBL process, this array of VO₂ nanocylinders was overlaid by a layer of bow-tie nanoantennas such that one nanocrystal was located precisely in the feed gap of each independently triggerable antenna, thus completing a metamaterial-perfect-absorber (MPA) configuration on top of a substrate consisting of a gold backplane covered with a thin layer of Al₂O₃ prepared by atomic-layer deposition. To test the modulation properties of the device, the temperature was varied from 21 °C to 87 °C, modulating the MPA resonance position from 1590 to 1230 nm, with a modulation depth of 37%, a switching speed of 1.5 ms and a switching energy of 21 nJ per pixel. Full-wave, finite-difference time-domain simulations indicate that this performance is roughly what should be expected. This performance substantially exceeds anything presently in the recent literature; the potential for electrical and optical modulation of the device will also be discussed.

Acknowledgements:

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Poster Abstracts

P1

High-speed highly-regular laser-induced periodic surface structures on metals: physical origin and prediction for a wide range of materials

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Highly-regular laser-induced periodic surface structures (HR-LIPSS) have been fabricated on surfaces of Mo, steel alloy and Ti, at a record processing speed on a large area and with a record regularity in the obtained sub-wavelength structures. The physical mechanisms governing LIPSS regularity are identified and linked with excited surface electromagnetic waves (SEWs). A material dependent criterion for obtaining HR-LIPSS is proposed for a large variety of metallic materials. The key irradiation parameters are determined for covering several cm² of material surface by HR-LIPSS in a few seconds while keeping the scanning direction misaligned relative to the laser polarization direction. Theoretical predictions suggest the possibility of HR-LIPSS production on principally any metal. This new achievement in the unprecedented level of control over the laser-induced periodic structure formation makes this laser-writing technology to be flexible, robust and hence highly competitive for advanced industrial applications based on surface nanostructuring.

Plasmonic initiated selective removal of thin Mo films by short and ultra-short lasers

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Mechanism of non-thermal patterning and selective removal of 40 nm Molybdenum (Mo) thin film from a Mo-Al-Mo hybrid structure is presented. The laser beam was focused using a galvo scanning system with a 100 mm focal length lens. The laser pulse overlap was controlled by adjusting the scan speed and the pulse repetition rate of the laser source. The position of sample and lens to target distance was precisely controlled by a 3D computer controlled stage. Atomic force microscope (AFM), scanning electron microscope (SEM), white light interferometer, and optical microscope were utilized for surface morphology and crater analysis.

For the case of infrared femtosecond laser interactions with Mo-Al-Mo, below the damage threshold fluence (0.33 J cm^{-2}), the absorbed laser energy led to nanostructure formation. The size of the conical pillar-like nanostructures are tenth of nanometres high and wide.

The origin of the nanostructures is considered in terms of electro-mechanical forces. Electrons energised by photon absorption, within one mean free path of the Mo surface, can in part migrate to more loosely bound surface states compared with those in the material and hence are more likely to escape from surface. The electric field in the vicinity of the surface leads to the expansion of the surface area to form a nano-pillars. The generation and pulling of nano pillars outwards from the surface, leads to clean and selective removal/delamination of Mo film below the recognised damage threshold fluence.

P3

High efficiency X-ray K α laser plasma source for medical imaging and material science

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Optimization of ultrafast x-ray emission from plasmas produced by femtosecond high intensity laser-solid interaction is still under strong investigation by many groups around the world. Motivation is related to important scientific applications like time-resolved x-ray diffraction or x-ray absorption fine structure spectroscopy for advanced high-resolution diagnostics of materials driven to extreme thermodynamic conditions and societal applications such as phase contrast imaging. Ultrashort K α X-ray pulses from laser plasmas are studied since decades but the continued improvement of femtosecond laser performances offers today the possibility to explore a wide range of regimes of laser interactions such as intensities $> 10^{19}$ W/cm 2 for pulse duration < 30 fs with controlled high contrast ratios using a unique laser source with moderate affordable peak power (~ 10 TW).

In the present work, Molybdenum K α line emission produced from ultrahigh intensity femtosecond laser solid interaction is experimentally studied over more than 2 orders of magnitude of temporal pulse contrast ratio as well as over a wide range of pulse intensity using the ASUR laser source [1]. The absolute yield of K α x-rays is measured as a function of contrast ratio from 6×10^{-8} to 3×10^{-10} and for a large intensity range 3×10^{16} - 4×10^{19} W/cm 2 . For intensity $I \geq 10^{19}$ W/cm 2 , we show that no saturation of the measured K α yield is observed. Furthermore, K α conversion efficiency, from laser pulse energy into total energy in K α line, is measured to be independent of intensity and reaches $\sim 2 \times 10^{-4}$ in 2π solid angle for all the studied contrast ratios.

The present results bring important information on different interaction regimes of high intensity femtosecond laser radiation with a solid target, since we observe a strong dependence of the K α emission on both laser pulse contrast and intensity. We will discuss of the interplay of different mechanisms like resonance absorption, vacuum heating (Brunel effect), plasma steepening by radiation pressure and JxB which are present at these different of interactions to explain our experimental results.

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Single temporally tailored femtosecond laser pulses for controlled high aspect ratio nanomachining of dielectrics

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We report on the generation of sub-wavelength channel structures with very high aspect ratios in fused silica with a single laser shot. To that end, femtosecond laser pulses with a wavelength centered around 785 nm are temporally tailored to match different electron excitation and recombination mechanisms at play. Laser pulses are focused above, on and below a fused silica surface under ambient conditions with a long distance microscope objective. Deep channels are created without making use of self-focusing and filamentation processes and analyzed by focused ion beam milling.

The channel depth as a function of focus position and pulse shape is reproduced by numerical calculations within a single parameter set. Channel depths in the range of several μm and diameters below 250 nm are demonstrated. Besides machining of nanophotonic devices in dielectrics, the technique has the potential to enhance laser based nano-cell surgery and cell poration techniques. [1]

References:

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P5

Temperature relaxation dynamics at the surfaces of metals and dielectrics upon ultrashort laser pulse irradiation in the regime of LIPSS formation

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In this work, to get insight into laser-induced periodic surface structures (LIPSS) formation, the relaxation of a modulation in the temperature profile is investigated numerically on surfaces of two different kinds of materials (metals and dielectrics) upon irradiation by an ultrashort laser pulse (~100 fs).

The temperature modulation is assumed to arise from the interference between the incident laser beam and the excited surface electromagnetic wave, which is considered here as the main mechanism at the basics of LIPSS formation [1]. The dynamics of dissipation of the temperature modulation toward the material depth is studied as it can provide an important information on the processes which occur upon LIPSS formation such as melting, ablation and stress development [2,3] and, thus, may bring a better control over the structures morphology.

The modelling is based on the two-temperature model. For dielectrics, the energy balance equations for lattice and electrons are supplemented with the rate equations for free-carrier generation by multiphoton absorption and collisional ionization and their relaxation by trapping into excitons. In both dielectrics and metals, the optical properties are taken into account through the Drude model with accounting for the evolution of the free-carrier density and/or temperature.

It is shown that strong temperature gradients can be formed along the surfaces of the materials under study within the fluence range of LIPSS formation in experiments. Considerable spatiotemporal variations in optical properties of the studied materials, including metals, are found as a feedback mechanism which must play a significant role in imprinting periodic structures on the material surfaces. The effects of the resulting nonlinear absorption on the surface temperature dynamics are discussed.

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Laser wavelength dependence on periodic surface structures induced by single-laser pulses

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Laser-induced periodic surface structures (LIPSS) are periodic modulation of the surface profile upon laser ablation with amplitudes and periodicity depending on the material properties and the laser ablation parameters. The formation of LIPSS is a single-step process of creating nanostructures on metals, semiconductor or dielectrics, providing new surface functionalities.

However, the physical origin of LIPSS is still under debate. Here, we present the formation of periodic surface structures induced by single-ultrashort laser pulses. Nanostructures have been defined by means of LIPSS with periods comparable to the laser wavelength. The dependence of the different laser parameters on the surface morphology is compared to the existing theories.

Accumulation effects in ultrashort-laser-induced air ionization: The role of metastable states of air molecules

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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 environmental effects on material processing by high-peak-power fs-laser pulses are still poorly understood due to a large variety and complexity of the involved processes. Here we report on a discovery of accumulation effects upon propagation of focused fs laser pulses (800 nm, 130 fs) in air under multi-pulse irradiation conditions. The transmission measurements have been carried out in air at different pressures and in vacuum for comparison. The spectroscopic measurements of air breakdown plasma have shown that the lines of molecular nitrogen emerge at fluences well below the breakdown threshold determined from the transmission measurements. The plasma absorption effects are found to be dependent on the pulse repetition rate and are considerably stronger at 1 kHz as compared to 10 Hz. This suggests that the metastable states of air molecules play an important role in initiation of air breakdown at high repetition rates. The spectroscopic data indicate that the ionization process involves the first triplet state of molecular nitrogen, $N_2(A^3\Sigma_u^+)$, with the life time of ~10 ms. Modelling of laser-induced air ionization shows a good agreement with the experimental data. The role of laser light absorption by air plasma at high-repetition-rate laser processing of materials will also be discussed.

Nonlinear detectors for dispersion compensation and pulse optimization in femtosecond laser material processing

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Femtosecond laser pulses are a unique tool for high precision processing of practically any kind of material. By using the technique of temporal pulse shaping, the optical breakdown in dielectrics can be manipulated to increase the efficiency of processing. We have shown that temporal Airy laser pulses are suitable to increase the precision of ablation in transparent dielectrics one order of magnitude below the optical diffraction limit [1, 2] and to generate channels with very high aspect ratios [3].

Especially when pulses shorter than 100 fs are used, the dispersion introduced by the optical components is important and its compensation is not a trivial task. Here we present our studies in using two- and three- photon photodiodes to compensate the dispersion in the interaction area of the process microscope and - in combination with the pulse shaper - to precisely measure the pulse duration by autocorrelation techniques.

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Laser amplification in laser excited dielectric materials

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Laser excitation of transparent dielectrics like sapphire or fused silica by ultra-short laser pulses usually cause a transition of the material to a transient metallic-like state. This is caused by the creation of a high carrier density in the conduction band, which is the first step in laser-material processing of dielectrics. This metallic state is typically identified by a high absorption and reflectivity as shown in a manifold of experiments.

In our recent studies, which are based on an in-line pump probe and a common-path spectral interference setup, we show that under appropriate conditions, the strong absorption in a thin laser excited dielectric sample is replaced by optical gain. The optical gain is coherent, directed and based on stimulated three-photon emission. We present energetic, temporal and spectral dependencies and properties of the laser excitation in a thin excited sapphire sample.

Synthesis of water-dispersible magnetic quantum dots - comparison between core- and shell doping

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Semiconductor quantum dots (QDs) are widely used as photostable, fluorescent dyes for bioimaging and diagnostic platforms. New synthesis approaches allow equipping QDs with magnetic properties in order to design biolabels that can be distinctively detected and localized through a combination of imaging techniques, e.g., employing fluorescence and magnetic resonance imaging. In this study, we present a facile aqueous synthesis procedure to manufacture either core- or shell-doped magnetic CdTe/ZnS QDs by using ferrous ions. Optical spectrometry, XRD and magnetic susceptibility measurements were used to compare the differences in their photoluminescence, structural and magnetic properties. The incorporation efficiencies of core- and shell-doped QDs were determined by HR-ICP-MS. Iron-doping of the QD's shell was about 1.4 times more efficient, and remarkably, the quantum efficiency of shell-doped QDs was about 1.7 times higher than for the core-doped ones. This could be explained by the fact that core-doping led to defects in the lattice and to a dramatic decrease in crystallinity, whereas shell-doping had no significant impacts on the crystal structure. This study also showed that water-dispersible, iron-doped core-shell QDs are applicable as bimodal contrast agents that show different temperature-dependent magnetic susceptibility, depending on whether iron was incorporated into the QD's core or its shell.

P11

Femtosecond laser material processing projects: Cell poration, starter-notches & LIBS and LIPSS

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We present an overview of three projects.

In cooperation with the GAP-Biophotonics group of the University of Geneva we enhanced the poration efficiency with a focused Gaussian laser beam in liquid environment by using temporal Airy pulses, instead of bandwidth limited pulses [1].

Our work with the Institute for Material Engineering in Kassel includes the production of well-defined starter-notches for crack investigations on the micrometer scale on preselected sites [2]. Cracks start at the tips of the notch and their growth can be explored. Furthermore, femtosecond laser-induced breakdown spectroscopy (fs-LIBS) was used for spatially resolved spectrochemical analysis of the surrounding material with μm -resolution [3].

Lastly, the investigation of laser induced periodic surface structures (LIPSS, “ripples”) has been an evergreen over the last few decades and there is still a lively discussion about the mechanisms behind LIPSS generation [4, 5]. We present large-number studies of LIPSS wavelength and area on fused silica and titanium.

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Apertureless scanning near-field nanolithography on metal and polymer films with a femtosecond Yb-doped fiber laser oscillator

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Optical nanolithography has experienced huge advances in the last decades. One of the most important trends is to reduce the size of the produced features, reaching nowadays spatial resolutions at the nanoscale. However, lithography resolution by optical methods has been limited by light diffraction limit. Apertureless scanning near-field optical lithography (aNFOL) can surpass this barrier [1-6]. In this method, a scanning probe microscope (SPM) tip brought down to a few nanometers from a substrate is illuminated by a focused femtosecond laser beam.

It is demonstrated that a non-amplified compact femtosecond Yb-doped fiber laser oscillator can well serve for this purpose [7]. The laser electromagnetic field is strongly enhanced at the tip-substrate gap, producing modifications at the surface of the substrate. The achieved resolution is controlled by the radius of the tip apex (5-10 nm). Thermal contributions are discussed based on heat accumulation [8]. Subwavelength surface structuring at the nanoscale is observed, with lateral resolution of about 10 nm and thus surpassing the light diffraction limit.

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P13

Femtosecond laser generation of microbumps on stacked Cu/Ag thin films

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Femtosecond ablation of metal thin films has been of vivid interest since decades [1,2]. Microbumps and nanojets have been observed on thin metal films below the ablation threshold [3,4]. The generation of microbumps has been attributed to two different mechanisms considering a loss of film strength without melting [5] and due to melting [3,4,6]. In order to resolve this contradictory issue, stacked copper/silver layers have been investigated in this study. The mixing of copper and silver is mainly restricted to the liquid phase. Thus, a mixing in the microbumps was taken as a criterion for the involvement of molten phases.

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**Merging spot size and pulse number dependence of femtosecond laser ablation thresholds:
Demonstration with, silicon, steel, and high impact polystyrene**

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The importance of femtosecond lasers as a tool in material machining applications is unquestionable. The energy density (fluence) required to modify a material is known as threshold fluence. Incubation meaning the repetitive pulsing and various irradiation area of the laser beam are two of the parameters influencing the threshold fluence. There are models in literature attempting to describe the dependence of threshold fluence on these experimental conditions. These models are mostly based on fitting the experimental data without a physical model behind and the mechanism of both phenomena is still quite unknown.

A new model describing both spot size and pulse number dependence of femtosecond laser ablation has been reported [1]. This model relies on attributing the mechanism of these phenomena on different types of defect in the material. Since defects strongly depend on material type, the model was demonstrated with various material classes: polymer, metal, semiconductor [2]. The new model can fit the experimental data and by that gives an insight into better understanding the underlying principle of femtosecond laser material processing.

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P15

Femto- and nanosecond pulse laser ablation dependence on irradiation area: role of defects in metals and semiconductors

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Modification thresholds are core parameters in laser materials processing as well as for optical components and telecommunication systems. The threshold fluence was found to be a material constant for a given set of experimental parameters such as pulse duration, wavelength, number of pulses and repetition rate. The modification threshold fluence however shows a dependence on the area that the laser pulse irradiates, both for femtosecond [1] and nanosecond [2] laser pulses. In literature, two models are known that describe the dependence of the threshold fluence on the spot size. They consider thermal accumulation [3] and defects densities [4].

In the present study [5], the irradiation area dependence of the modification behavior of steel, and silicon with femtosecond and nanosecond pulsed lasers are investigated for various spot sizes, and intentionally increased defect densities. The beam radius region, where there is a transition from intrinsic to defect-mediated ablation phenomena, shifts to smaller beam radii for higher defect densities. This confirms the role of defects in irradiation area dependence of ablation thresholds.

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Influence of laser irradiation area and pulse number on top-down (ablation) and bottom-up (LIPSS) processes

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The mechanism of the laser-induced periodic surface structure (LIPSS, "nano-ripples") formation is still controversially discussed in literature. Several mechanisms have been proposed, such as interference effects along with transient changes in the optical properties during laser irradiation [1], second harmonic generation in compound semiconductors [2], excitation of surface excited waves [3], or self-organization [4].

In a recent study [5-8], we could describe the reduction of laser ablation threshold fluence F_{th} with pulse number N and beam radius w . The reduction of F_{th} with w could be related to the coverage of low-density defects (LDDs) whereas incubation, i.e. the reduction of F_{th} with N is attributed to high-density defects (HDDs).

Here, we report on the role of N and w for LIPSS formation on silicon and stainless steel. On silicon, irradiation with few pulses ($N \leq 5$) generates low spatial frequency LIPSS (LSFL) only for beam radii greater than the mean separation of LDDs ($w \geq 50 \mu\text{m}$). Higher pulse numbers ($N > 5$) produce LSFL and high-spatial frequency LIPSS (HSFL) independent of the beam size. For stainless steel, on the other hand, HSFL and LSFL are also found from the first pulse on for $w \geq 10 \mu\text{m}$. This is attributed to the higher number of LDDs of stainless steel compared to silicon.

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