9th European Conference on Applications of Femtosecond Lasers in Materials Science

FemtoMat 2022

March 14 - 16, 2022
Mauterndorf Castle, Mauterndorf
Salzburg, Austria
https://esg-nano.ac.at/femtomat

Chair: Wolfgang Kautek
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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,

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

the 7th FemtoMat 2017, Mauterndorf, Salzburg, Austria, March 2017,

and the 8th FemtoMat 2019, Mauterndorf, Salzburg, Austria, March 2019, is the basis of the present

9th FemtoMat 2022, Mauterndorf, Salzburg, Austria, March 14 - 16, 2022.

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 2022

Wolfgang Kautek

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Program

Monday, March 14, 2022

08:30 – 09:00 **Registration**
09:00 – 09:15 **Opening and greetings**
09:15 – 09:45 **L01** Leonid Zhigilei (University of Virginia, USA)
Computational study of nonlinear propagation of laser-induced acoustic waves and acoustic activation of surface processes
09:45 – 10:15 **L02** Nadezhda Bulgakova (HiLASE Centre, CZ)
Modeling of ultrafast laser interaction with silicon: Critical assessment of excited processes through comparison of simulations with experiment
10:15 – 11:45 **Posters:** Short lectures and session (with coffee)
11:45 – 12:15 **L03** Bärbel Rethfeld (TU Kaiserslautern, DE)
Role of surface plasmon polaritons in ultrafast nanostructuring and nanophotonics

12:15 – 17:00 **Free discussion**

17:00 – 17:15 **Coffee**
17:15 – 17:45 **L04** Walter Pfeiffer (University of Bielefeld, DE)
Attosecond dynamics in solids after photoexcitation as a benchmark for our understanding of complex quantum systems
17:45 – 18:15 **L05** Thibault Derrien (HiLASE Centre, CZ)
Ultrafast excitation of electrons in crystals: insights from non-equilibrium band structure calculations
18:15 – 18:45 **L06** Jens Biegert (ICFO, ES)
Real-time interaction of photons, carriers and lattice with attosecond soft X-ray spectroscopy
18:45 – 19:00 **Coffee**
19:00 – 19:30 **L07** Goran Erik Hallum (HM Hochschule München University of Applied Sciences, DE)
Dynamics of ultrafast indium tin oxide ablation
19:30 – 20:00 **L08** Martin Aeschlimann (TU Kaiserslautern, DE)
Ultrafast Surface Science with Light carrying Orbital Angular Momentum
20:00 – 20:30 **L09** Wolfgang Kautek (University of Vienna, AT)
Femtosecond laser generation of binary metallic oxide nanoparticles for applications in medical imaging diagnosis

Tuesday, March 15, 2022

09:00 – 09:30 **L10** Razvan Stoian (Université Jean Monnet, FR)
Non-diffractive ultrafast laser beams for advanced material processing
09:30 – 10:00 **L11** Gert-Willem Römer (University of Twente, NL)
Ultrashort pulsed laser processing of sapphire and subsequent isotropic and anisotropic wet chemical etching
10:00 – 10:30 **L12** Cyril Mauclair (Université Jean Monnet, FR)
Ultrafast laser surface and bulk processing: the interest and challenges of spatial beam control and in-situ characterization
10:30 – 10:45 **Coffee**
10:45 – 11:15 **L13** Alexander Bulgakov (HiLASE Centre, CZ)
Dual wavelength femtosecond laser-induced damage and ablation of silicon
11:15 – 11:45 **L14** Nicolas Sanner (Aix-Marseille Université, FR)
Processing arrays of deep nanoholes at the surface of glass by short-length micro-Bessel beams
11:45 – 12:15 **L15** Jan Gerrit Horstmann (Max Planck Institute of Multidisciplinary Sciences, DE)
Ultrafast probing and coherent vibrational control of surface structural phase transitions

12:15 – 17:00 **Free discussion**

17:00 – 17:15 **Coffee**
17:15 – 15:45 **L16** Emmanuel Stratakis (IESL - FORTH, GR)
Ultrafast laser fabrication of biomimetic surfaces
17:45 – 18:15 **L17** Francois Courvoisier (FEMTO-ST, FR)
Resonance absorption and second harmonic generation on laser-induced nanoplasmas
18:15 – 18:45 **L18** Jan Hrabovsky (HiLASE Centre, CZ)
Formation of tubular structures and microneedles on silicon surface by doughnut-shaped ultrashort laser pulses
18:45 – 19:00 **Coffee**
19:00 – 19:30 **L19** Stefan Haessler (ENSTA Paris, FR)
Ultra-high intensity laser-driven plasma mirrors as light and particle sources
19:30 – 20:00 **L20** Johannes Heitz (Johannes Kepler University Linz, AT)
Micro- and Nanopatterning of Bionic Materials
20:00 – 20:30 **L21** Aleksandr Ovsianikov (TU Wien, AT)
High Resolution 3D Printing and Bioprinting with Femtosecond Lasers

Wednesday, March 16, 2022

09:00 – 09:30 **L22** Peter Banzer (TU Graz, AT)
Nanoscale Structured Light-Matter Interactions - Unique Applications and Promising Opportunities

09:30 – 10:00 **L23** János Hebling (University of Pécs, HU)
Ultrashort THz pulse generation and time resolved measurements in the THz range on solids and biological materials

10:00 – 10:30 **L24** X. Sedao (Université Jean Monnet, FR)
Ultrafast Laser Micro and Nanoprocessing for Biomedical Applications

10:30 – 10:45 **Coffee**

10:45 – 11:15 **L25** Maximilian Spellauge (HM Hochschule München University of Applied Sciences, DE)
Comparison of the ablation dynamics of gold immersed in air and water by pump-probe microscopy

11:15 – 11:45 **L26** Peter Balling (Aarhus University, DK)
Ultrashort-pulse laser excitation of dielectric materials: New knowledge from thin samples?

11:45 – 12:15 **L27** Nikolaos Giannakaris (Johannes Kepler University Linz, AT)
Femtosecond Single-Pulse and Orthogonal Double-Pulse Laser-Induced Breakdown Spectroscopy (LIBS): Femtogram Mass Detection and Chemical Imaging with Micrometer Spatial Resolution

12:15 – 17:00 **Free discussion**

17:00 – 17:15 **Coffee**

17:15 – 17:45 **L28** Stella Maragkaki (IESL - FORTH, GR)
LIPSS on pre-patterned surfaces

17:45 – 18:15 **L29** David Redka (HM Hochschule München University of Applied Sciences, DE)
The role of early mechanical surface expansion for the pulse duration dependency of the ablation efficiency in the CrMnFeCoNi high-entropy alloy

18:15 – 18:45 **L30** Cristian Sarpe (University of Kassel, DE)
Identifying malignant tissue using fs-LIBS and machine learning algorithms

18:45 – 19:00 **Coffee**

19:00 – 19:30 **L31** Markus Huber (University of Regensburg, DE)
Femtosecond nano-videography of charge carrier dynamics in atomic-scale systems

19:30 – 19:45 **Closing remarks**

Lecture Abstracts

L01

Computational study of nonlinear propagation of laser-induced acoustic waves and acoustic activation of surface processes

Maxim V. Shugaev, Yuan Xu, Leonid V. Zhigilei

University of Virginia, USA

The ability of laser-induced surface and bulk acoustic waves to substantially enhance the rates and selectivity of heterogeneous catalytic reactions, desorb molecules, and facilitate surface diffusion of atomic clusters, has been demonstrated in a number of experimental studies. Despite the intriguing nature and potential practical importance of these observations, the theoretical efforts aimed at interpretation of these results has been limited, and the understanding of the fundamental mechanisms and elementary processes responsible for the acoustic activation of surface processes is still far from being complete.

In the study reported in this presentation, we apply multiscale modelling to the exploration of the mechanisms that may be responsible for increased catalytic activity of surfaces subjected to surface and bulk acoustic waves. A series of continuum-level simulations is first applied for the investigation of the generation of strong acoustic waves by short pulse laser irradiation. The irradiation parameters, such as laser fluence, pulse duration, and spot size are optimized for the generation of acoustic waves of suitable strength and frequency range. Large-scale molecular dynamics simulations are then used for the exploration of the ability of strong nonlinear acoustic pulses to (1) induce desorption of atomic clusters from surfaces, (2) cause exfoliation of graphene multilayers from a Cu (111) substrate, and (3) produce dislocations in the surface region of substrates. The emission of dislocations can result in the formation of atomic steps on the surface and may affect its catalytic activity, while the acoustic desorption can have important implications for various fields ranging from cold desorption of heat-sensitive molecules for mass spectrometry analysis to the removal of products of chemical reactions and regeneration of catalytic activity of metal surfaces. The computational predictions are related to the available experimental data.

L02

Modeling of ultrafast laser interaction with silicon: Critical assessment of excited processes through comparison of simulations with experiment

N.M. Bulgakova, A.V. Bulgakov

HiLASE Centre, Institute of Physics of the Czech Academy of Sciences, Dolní Břežany, Czech Republic

Laser processing of material surfaces is one of the key technologies for state-of-the-art novel applications in micro- and nanoelectronics, photonics, sensing technologies, photovoltaics, and in other fields. Silicon as one of the most abundant materials whose importance in scientific and industrial applications is not diminishing with time. Laser treatment of this material is an extremely complicated phenomenon, which proceeds through triggering of a wealth of linear and nonlinear, thermal and nonthermal processes. Many numerical models have been developed targeting on prediction of optimal regimes of silicon processing with short and ultrashort laser pulses. However, most of the models are usually limited to an only laser wavelength or one pulse duration with the use of a number of fitting parameters and their direct application for other laser irradiation parameters often does not provide agreement with experimental data.

In this report, we will analyze step by step all the processes involved in silicon excitation and heating by pico- and femtosecond laser pulses at 1030 nm wavelength. This involves single- and multiphoton absorption, avalanche ionization, free electron recombination, dynamic optical response of the laser-excited surface layer, melting threshold and melting dynamics at over-threshold laser fluences. The analysis will be performed based on direct comparison with the experimental data on the ablation thresholds and crater depths obtained by using a PHAROS laser (Light Conversion, 1030 nm) at pulse durations of 260 fs and 7 ps. A critical assessment is given of the use of the concepts of plasma screening in Auger recombination [1], electron density dependence of energy coupling to the lattice and the hot carrier relaxation time [2], applicability of the multi-layer reflectivity model [3,4], ambipolar diffusion [5-7], and two-photon absorption coefficient as an effective value for several nonlinear absorption processes [7,8]. Finally, a set of silicon parameters is outlined, which yields a good agreement between the modeling and experimental results for 1030 nm wavelength, which is widely used for material processing.

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L03

Role of surface plasmon polaritons in ultrafast nanostructuring and nanophotonics

Baerbel Rethfeld, Pavel N. Terekhin

Department of Physics and Research Center OPTIMAS, Technische Universität Kaiserslautern

Surface plasmon polaritons (SPP) can be excited at a step edge of a metallic surface. The interference of the laser with the SPP can lead to a spatial modulation of the energy within the material [1]. We reveal different cases where this modulated energy input has an influence on measurable quantities. We compare theoretical considerations with experimental measurements for low and high excitation regimes.

For weak excitation, the decay of SPP into hot carriers is important for nanophotonic applications. Via spatiotemporal separation, the energy and momentum distribution of surface plasmon-induced hot carriers can be isolated [2].

For strong excitation, we have studied the role of SPP on nanostructuring, namely the formation of laser-induced periodic surface structures (LIPSS) near a predesigned gold step edge following single-pulse femtosecond laser irradiation [3]. This work includes atomistic-continuum simulations as well as experiments detecting nanosized periodic surface features. It reveals two key components of LIPSS formation: SPP excitation, responsible for laser-deposited energy modulation, and material reorganization, responsible for the final LIPSS morphology. We find an excellent agreement in the periodicity and the decay of LIPSS between experimental results and numerical simulations [3].

[1] P.N. Terekhin, O. Benhayoun, S.T. Weber, D.S. Ivanov, M.E. Garcia, B. Rethfeld, Influence of surface plasmon polaritons on laser energy absorption and structuring of surfaces, Appl. Surf. Sci. 512, 144420 (2020).
[2] M. Hartelt, P.N. Terekhin, T. Eul, A.-K. Mahro, B. Frisch, E. Prinz, B. Rethfeld, B. Stadtmüller, and M. Aeschlimann, Energy and Momentum Distribution of Surface Plasmon-Induced Hot Carriers Isolated via Spatiotemporal Separation, ACS Nano 15, 19559 (2021).
[3] P.N. Terekhin, J. Oltmanns, A. Blumenstein, D.S. Ivanov, F. Kleinwort, M.E. Garcia, B. Rethfeld, J. Ihlemann, and P. Simon, Key role of surface plasmon polaritons in generation of periodic surface structures following single-pulse laser irradiation of a gold step edge, Nanophotonics 11, 359 (2022).

L04

Attosecond dynamics in solids after photoexcitation as a benchmark for our understanding of complex quantum systems

Walter Pfeiffer

Bielefeld University

In recent years, time-resolved spectroscopy of electron dynamics in solids advanced to the attosecond regime, i.e., on the time scale on which electron motion occurs on an atomic scale. In such experiments an extreme ultraviolet (EUV) pulse with durations of typically a few 100 as is used to excite the solid and the response is probed by correlating this excitation with a field in the near infrared (NIR). One example of such a technique is attosecond time-resolved streaking spectroscopy [1]. This technique allows investigating relative temporal delays in the photoemission from different initial states. Discrepancies between experimental observations and existing theoretical models challenge our understanding of the photoemission process.

Using this technique, we were recently able to demonstrate that the present theoretical model of solid-state photoemission misses an essential effect that significantly affects such photoemission delays [2]. Intra-atomic delays determined by the involved angular momentum of the photoelectron affect the photoemission times and are not accounted for in present theoretical models of solid-state photoemission.

In our present approach to describe the observations we dissect the photoemission process artificially in an intra-atomic and a propagation part that are modeled based on theoretical concepts from atomic physics and solid-state physics, respectively. To overcome this artificial dissection new theoretical models of the photoemission process are needed and, hence, attosecond time-resolved spectroscopy provides new benchmarks for a further development of a complex quantum system, i.e., the dynamics of the many-body problem of a photoelectron interacting with the remaining photohole and the other electrons.

[1] A. L. Cavalieri, N. Müller, T. Uphues, V. S. Yakovlev, A. Baltuska, B. Horvath, B. Schmidt, L. Blumel, R. Holzwarth, S. Hendel, M. Drescher, U. Kleineberg, P. M. Echenique, R. Kienberger, F. Krausz, U. Heinzmann, Attosecond spectroscopy in condensed matter. Nature. 449, 1029–1032 (2007).

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L05

Ultrafast excitation of electrons in crystals: insights from non-equilibrium band structure calculations

Thibault J.-Y. Derrien

HiLASE Centre - Institute of Physics

Last decade, understanding of transient excitation of electrons in solids has brought important developments for several classes of materials, at the level of both fundamentals and applications. While laser-excitation of dielectrics induces measurable ultrafast currents in the PHz regime [1], employing few-cycle laser pulses with controlled carrier envelope phase enables a coherent control of the electron dynamics with reduced crystal damage probability [2,3]. Notably, the possibility of transiently closing the band-gap of solids during their irradiation by linearly polarized ultrashort laser pulses was evidenced and attributed to the light-induced Zener tunneling [3,4].

The corresponding ultrafast modification of the band structure induced by laser dressing of electronic states can be measured experimentally [5,6] and analyzed theoretically using the Floquet formalism [7,8]. Despite its simplicity and limitations, it is applicable to several classes of materials [5,6,8] and enables to study the effects of light coupling with electrons in solids for a wide range of experimental conditions [9].

In this work, after preparing the electronic band structures of two metals (Au and Mo), a semiconductor (Si) and a dielectric (α-SiO_2) using the density functional theory (DFT), the effects of dressing by a polarized laser light on the corresponding electronic band structures were investigated using the Floquet formalism [9]. While a selective excitation of the electrons can be achieved via a choice of laser wavelength and field strength [9], the Floquet simulations illustrate how the change in crystal orientation [10-13] affects the electron dynamics in solids.

Overall, the proposed approach outlines promising ways for selecting materials and laser parameters, via a computer-aided manner, broadening perspectives in ultrafast photonics [5].

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L06

Real-time interaction of photons, carriers and lattice with attosecond soft X-ray spectroscopy

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The conversion of light to fundamental excitations of matter is governed by the build-up of electronic coherences and their dephasing to excited quasiparticles due to scattering processes, which occur on atto- and femtosecond timescales. Disentangling the interplay of these mechanisms, and how they lead to a specific flow of energy inside a material, is extremely challenging since many of these effects occur on overlapping temporal scales. I will discuss the semimetal graphite, which was investigated with attosecond K-shell X-ray absorption near edge structure (XANES) spectroscopy and show how the combination of our new measurement methodology with theoretical modelling allows to assign the spectroscopic signatures to microscopic processes relating to the dynamic evolution of electrons, holes and phonon modes of the material. Lastly, since this new method is generally applicable to molecules and solids, I will point out some possibilities to address canonical questions relevant for light-harvesting and general non-equilibrium multi-body physics of phase-transitions.

L07

Dynamics of ultrafast indium tin oxide ablation

Goran Erik Hallum[1], Dorian Kürschner[2], Wolfgang Schulz[2], Heinz Paul Huber[1]

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Indium tin oxide (ITO) films are commonly used as transparent electrodes for various applications such as touch displays, LEDs, or solar cells. Selective removal of material is a necessary process step for many ITO applications that stands to be optimized by ultrafast laser ablation. We observe an ablation threshold fluence for ITO thin films on float glass substrates to be an order of magnitude less than the ablation threshold for glass substrates, providing the desired selectivity for industrial processing. 100 nm thick ITO films on float glass display an ablation threshold for peak fluences of approximately 0.2 J/cm² whereas glass substrates of float glass or quartz display thresholds of approximately 3 or 4 J/cm², respectively, for pulse durations of 525-700 fs at a wavelength of 1056 nm and a beam waist radius of 15 µm.

Optimized ultrafast laser structuring parameters exist in literature, but unwanted effects still exist such as burr, droplet, and crack formation. The ablation mechanism is furthermore seen to be dependent on a variety of laser parameters. In our study, we seek to understand the underlying mechanisms for ablation given a wide range of industrially available laser parameters. Only after the influence of these parameters on the ablation mechanism is understood using pump-probe microscopy, can the process be reliably and reasonably improved.

We have investigated the ablation dynamics of thin film ITO on glass substrates investigated using pump-probe reflectometry from picosecond to microsecond delay times [1]. An initial sharp rise in reflectivity was observed as a result of free electron generation with a decay time of a few picoseconds followed by a drop in reflectivity attributed to a phase explosion, a direct ablation process, for fluences above the ablation threshold for tens of picoseconds. A rate equation system is used to further examine the free electron dynamics, where it is seen that photoionization is primarily achieved through impact ionization. Photomechanical film bulging was then observed in the form of Newton's rings for delay times of hundreds of picoseconds, giving evidence for a confined energy situation. For nanosecond time delays, particle generation was seen at the edge of the irradiated spot, which gave way to a radially expanding particle cloud for tens of nanoseconds. For fluences above 0.7 J/cm², a second bulging event was observed after one nanosecond. Here it is seen that multi-pulse processing is required to efficiently remove the entirety of the film.

As an extension to the examination of ITO ablation in air, we examine substrate irradiation, where the laser pulse is incident from glass focused on the interface between ITO and glass. Direct comparisons are made to the system in air. A defect-rich layer at the ITO-glass interface, which has been seen to be highly electrically resistive [2], is seen to have an impact on the electron dynamics. For fluences near the ablation threshold, we observe that the material above this electrically "dead" layer is preferentially removed. For an increase in pulse energy, we observe the complete removal of the ITO layer at the bottom of the crater. This is followed by island formation for a further increase in the pulse energy, similar to that seen in Si_3N_4 [3]. In pump-probe microscopy, we observe a characteristic darkening of the reflectivity associated with the optical breakdown in glass. Here we see a preferential non-linear absorption in glass after laser intensities cross a critical threshold. For ultrafast laser scribing applications, it is seen that excessive energy is not only unnecessary, but also leads to the damage of the substrate material. Understanding that the bottom 15-20 nm of the ITO layer is not electrically conductive allows us to further reduce the necessary fluence for scribing applications using substrate irradiation.

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L08

Ultrafast Surface Science with Light carrying Orbital Angular Momentum

Martin Aeschlimann

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Optical fields can carry orbital angular momentum (OAM) in helical beams with an azimuthal phase dependence. Since its discovery in 1992 by Allen et al [1], a variety of applications for the OAM of light have been developed. Optical OAM, like spin, can be transferred to particles in optical tweezers and rotate them mechanically [2]. More recently, transfer to bound electrons has been demonstrated for a quadrupole transition in a trapped calcium ion [3] and for solids such as MoS_2 [4]. In addition to its contribution to fundamental research, the OAM of light is also very promising for a variety of applications such as data transmission, micromanipulation, imaging, and quantum information technology.

In my talk, I will focus on exploring the potential impact of the OAM of light on laser-induced ultrafast surface science and magnetism. I will explain the underlying physical mechanisms of light with OAM, introduce various methods for its generation, and present two selected applications in detail.

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[2] H. He et al., Phys. Rev. Lett. 75, 826 (1995).
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L09

Femtosecond laser generation of binary metallic oxide nanoparticles for applications in medical imaging diagnosis

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3 Departamento de Física de La Materia Condensada and Institute of Research on Electron Microscopy and Materials (IMEYMAT), Universidad de Cadiz, Puerto Real (Cadiz), Spain
4 Instituto de Nanociencia y Materiales de Aragón (CSIC-University of Zaragoza), Zaragoza, Spain
5 Photonics Institute, Technische Universität Wien, Vienna, Austria
6 Instituto de Cerámica y Vidrio (CSIC), Madrid, Spain

Liquid-Assisted Pulsed Laser Ablation (LA-PLA) is as a promising alternative top-down method to directly synthesize colloidal dispersions of nanoparticles (NPs), because of its low cost, simplicity, and environmental friendliness [1,2]. LA-PLA is particularly attractive for medical applications since the use of stabilizing toxic molecules can be avoided and chemical precursors are replaced by less costly bulk raw materials [3]. Contrast enhanced digital mammography and magnetic resonance imaging in combination with X-ray film mammography in breast cancer screening are complementary. A multimodal approach can be performed without significant delay during the same imaging session. It requires the development of multimodal imaging contrast agents (MCAs) capable of responding to both X-ray radiation and magnetic fields [3-7]. The presented work was to design and optimize the fabrication of nanoparticles for MCA applications with sub-30 fs LA-PLA. The laser targets were ceramic samples combining a magnetic iron oxide, such as γ-Fe_2O_3 or Fe_3O_4, and an oxide phase of a radiopaque metallic element with high atomic weight, such as W, Ta and Bi.

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L10

Non-diffractive ultrafast laser beams for advanced material processing

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Material structuring represents today the base for rapidly growing application areas in emerging technologies. Ultrafast lasers contribute essentially to the development of micro/nanotechnologies, being able to structure materials with utmost precision. Alongside efficiency, the question of resolution is crucial for further industrial insertion and the challenge lies now in pushing the capacity to reach the nanoscale, with resulting morphologies defining upgraded functions to be attached to the structured material. Optical processing being typically limited by diffraction to scales comparable to the wavelength, an alternative strategy to achieve extreme scales relies on harvesting material reactions to light, experiencing thus not an optical limit but a material organization limit which, for a large range of material phenomena, lies one order of magnitude below. We will discuss ultrafast laser irradiation concepts, combining beam engineering in space and time, capable of achieving structuring features with sub-wavelength characteristic sizes in direct irradiation modes as well as in periodic self-organization processes. We indicate as well significant advantages of non-diffractive beams to structure materials with unprecedented resolution as well as with high efficiency. These advantages pertain to both transparent and non-transparent matter. A range of applications will be discussed.

L11

Ultra-short pulsed laser processing of sapphire and subsequent isotropic and anisotropic wet chemical etching

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Crystalline sapphire (α-Al_2O_3) is a transparent material characterized by a high melting temperature (2300 K), high hardness (9 Mohs scale), a low friction coefficient and is a good thermal and electrical insulator. It is also inert to many wet chemicals. Due to these properties, sapphire is used in e.g., semiconductor industry, microfluidics, smartphones, watches and as a substrate in integrated photonic devices. However, especially because of its hardness, it is difficult to machine with conventional manufacturing processes, such as mechanical dicing or milling. As sapphire is transparent to light with wavelengths in the range of about 0.3 µm to about 4 µm, a laser beam can be focused below its surface, in the bulk. Focusing a femto- or picosecond pulsed laser beam with intensity levels over 10^{14} W/cm^2, inside sapphire, results in an amorphized volume (with dimensions in the range of µm's) in and near the laser focus. In a subsequent step, this amorphized sapphire can be removed selectively by chemical wet etching. This two-step method (laser processing and etching) results in hollow structures inside the substrate [1]. Not only isotropic etchants (e.g., hydrofluoric acid), but also anisotropic etchants (e.g., a mixture of sulphuric acid and phosphoric acid) can be used to etch sapphire. The first etchant mainly reacts with the amorphized sapphire, whereas the indicated anisotropic etchant also etches the crystalline material, but at a much lower rate. We combine an experimental and a theoretical approach study and optimize this two-step method. Our numerical model allows simulation of the laser-material interaction (absorption and energy relaxation) during short pulsed laser processing of sapphire [2]. From simulations it is concluded that avalanche ionization needs to be triggered for sapphire to significantly absorb laser energy. From electron microscopy analysis of cross-sections of processed samples, we conclude that the laser focus position (either below or on the surface of the substrate) and the laser pulse energy are the most dominant laser parameters determining the morphology of the laser-induced amorphized zones [1,3]. Further, from experimental results we conclude the type of etchant used for the study has a strong effect on the resulting structures, not only in the bulk, but also on the surface of sapphire, and that intricate bulk and surface features can be obtained which can be exploited in the above-mentioned applications.

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L12

Ultrafast laser surface and bulk processing: the interest and challenges of spatial beam control and in-situ characterization

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Laboratoire Hubert Curien

Ultrafast laser pulses can be used to achieve surface and bulk processing with micro-nanometer scale by direct focusing of the sample surface of inside the transparent medium. With the reduced amount of thermal effects, a high structuring precision can be achieved. The technique is gaining interest for industrial as well as biomedical applications as nowadays ultrafast laser sources are more and more reliable, with performances allowing for high repetition rate processing. In this trend, any technique that can reduce the laser processing time and better control the interaction zone can be game changing. In this contribution, both of these topics are covered. First, advances in spatial beam shaping for surface and bulk processing are discussed, particularly in the context of cataract surgery where a change of paradigm has recently occurred to achieve full emulsification of the crystalline lens.

On the other side, in-situ monitoring of the interaction zone appears to be the next challenge for smart processing. In particular, a technique to follow the pulse-to-pulse growth of Laser Induced Periodic Surface Structures (LIPSS) based on structured illumination is used for in-situ observation of the irradiated surface. While the technique permits observing interesting phase locking mechanisms, the application potential of such optical super resolution techniques is underlined for large surface process monitoring.

L13

Dual wavelength femtosecond laser-induced damage and ablation of silicon

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To date, there exist a limited number of studies on bi-chromatic laser irradiation of materials demonstrating or predicting significant advantages of these regimes, at certain conditions, for laser applications including enhancement of the processing efficiency [1-3], higher nanoparticle yields [4] and improved quality of surface nanostructuring [5]. The bi-chromatic irradiation regimes are still poorly investigated and we have performed a detailed experimental and theoretical study on laser ablation and surface modification of monocrystalline silicon by dual-wavelength femtosecond irradiation using the fundamental (1030 nm) and second harmonics (515 nm) outputs of a PHAROS laser (260 fs pulse duration). The spot sizes of both pulses at the surface were adjusted to be identical to avoid uncertainty in fluence determination. The morphology of the produced spots is investigated by microscopy methods as a function of the total laser fluence (in the range 0.2 – 2 J/cm^2), time delay between the pulses (in the range from -100 ps to +100 ps, where the negative delay corresponds to conditions when the 515 nm pulse comes first), and the ratio of the pulse energies. The results are compared with those obtained in monochromatic irradiation regimes. Theoretical modeling has been performed to describe bi-color laser-induced excitation, heating and melting of the irradiated Si target under the experimental conditions.

We demonstrate that a combination of IR and visible fs-laser pulses at some negative time delays can result in a more efficient ablation of silicon with a better crater quality as compared to monochromatic irradiation. The damage area is maximal at zero-time delay. The observations are in line with the model calculations. The results provide insight into features and mechanisms of dual-wavelength laser processing offering a better control of the energy deposition into material.

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L14

Processing arrays of deep nanoholes at the surface of glass by short-length micro-Bessel beams

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Arrays of holes with subwavelength diameters and depths of several micrometers constitute the building block of devices that open attractive applications like 2D photonic crystals, 2D metamaterials or nanostructured surfaces. Here we present a technique based on picosecond laser processing with customized beam shaping, allowing to bridge the gap between the current capabilities of fabricating on one hand nanoholes but of shallow depths (<< 1 µm) – by regular Gaussian beam ablation – and on the other hand very long nanochannels (>> 100 µm) – by Bessel beam shaping.

To this aim, we developed a beam shaping methodology that offers a new degree of freedom to Bessel beams: the control of their length, irrespective of their core diameter. Then we use these short-length micro-Bessel beams to machine square arrays of nanoholes (typically ~200-300 nm diameter) with variable depths (ranging from 3 µm to 10 µm) by direct laser ablation at the surface of fused silica, as a prospective demonstration of the potential of this method towards the realization of photonic integrated devices.

L15

Ultrafast probing and coherent vibrational control of surface structural phase transitions

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Solid-state surface systems are particularly attractive because of their modified electronic, lattice and spin structures, resulting in strongly altered physical and chemical properties compared with the bulk [1]. We have recently developed ultrafast low-energy electron diffraction (ULEED) in a laser pump/electron-probe scheme to explore optically-induced structural dynamics at surfaces on their intrinsic time scales [2,3,4].

This talk will introduce the basic principles of ULEED and discuss our recent advances regarding the observation and control of structural transitions far from equilibrium, involving, e.g., the transfer of concepts from femtochemistry to solids. In this context, we report the coherent vibrational control over the phase transition in a quasi-one-dimensional Peierls insulator by manipulating the vibrational amplitudes of key lattice modes [5]. Specifically, we employ optical multi-pulse excitation schemes and monitor the structural transformation between the insulating (8×2) and the metallic (4×1) phase of atomic Indium wires on Si(111) by ULEED. An analysis of the delay-dependent phase transition efficiency proves the distinct roles of shear and rotation phonons for the transition and provides insight into the underlying control mechanisms and structural pathways. Mode-selective excitation of shear and rotation modes can further be used to control the trajectory of the system across the transition state on the excited potential energy surface.

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L16

Ultrafast laser fabrication of biomimetic surfaces

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The fabrication of artificial biomimetic surfaces via femtosecond laser processing is presented. Metallic, semiconductor and dielectric surfaces were irradiated and Laser Induced Surface Structures (LIPSS) were observed in each type of material. In particular, femtosecond laser pulses with linear, circular, radial or azimuthal polarization states were used for structuring steel, silicon and fused silica surfaces. Experimental results showed that the direction of LIPSS in each case proved to be dependent on the laser beam polarisation. A complete study was carried out for the investigation of LIPSS dependence on fluence and the number of pulses per spot for variable beam polarization states and irradiation strategies, allowing the production of new and more complex surface structures. Furthermore, we present a novel way to control and modulate the different LIPSS morphologies and geometries. Moreover, large area surfaces were fabricated, tailored with various micro-and/or nano- structures bearing great structural resemblance to surfaces found in nature such as the lotus leaf, shark skin and butterfly Greta Oto wing. Those bioinspired surfaces manifest remarkable optical and wetting properties, which were attributed to the specific surface morphology. Thus, femtosecond laser processing can be a novel and one single-step method for the fabrication of functional surfaces on almost all classes of solid materials.

L17

Resonance absorption and second harmonic generation on laser-induced nanoplasmas

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Spatial shaping of laser pulses into Bessel pulses allows for generating nano-plasmas in the bulk of dielectrics with plasma profiles that are much steeper than those conventionally produced using Gaussian beams. With experiments and first-principles numerical simulations, we demonstrate the generation of over-critical plasma densities on long distances within sapphire. Collisionless resonance absorption and generation of second harmonic are identified within the materials bulk.

L18

Formation of tubular structures and microneedles on silicon surface by doughnut-shaped ultrashort laser pulses

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We report on the formation of tubular structures and microneedles on the surface of monocrystalline silicon using ultrashort laser pulses. Highly deterministic surface processing is ensured by single-shot ablative modification of the sample surface using radially polarized doughnut-shaped laser pulses with duration of 70 fs. Under such conditions, well reproducible tubular structures are formed whose height is rising with increasing fluence, culminating by closing the structure on the top with formation of a microneedle with a cavity in its base. Upon multi-pulsed irradiation, the height of the needle structures can further increase as compared to those produced by single pulses and top part of the structure is flattened. However, at a certain number of pulses, melting and ablation cause collapsing the entire structure. The mechanisms responsible for creating the tubular and needle structures are discussed based on the careful analysis of the experimental observations. The generated silicon microtubes and needles can find applications in various field, such as intracellular delivery of micromolecules, micro/electromechanical systems, photovoltaic devices and silicon-based photonics.

L19

Ultra-high intensity laser-driven plasma mirrors as light and particle sources

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Plasma mirrors are overdense plasmas created at the surface of laser-ionized optically flat solid targets. If the surface plasma density gradient is sufficiently steep, they behave as high-quality mirrors, which turn into versatile nonlinear optical devices when driven by ultra-intense femtosecond laser beams ($I > 10^{16}$ W/cm^2). They are recognized as an excellent model system for ultra-high intensity laser-plasma interactions and as promising secondary sources of intense attosecond light pulses through high-order harmonic generation, as well as for laser-driven electron and ion acceleration.

We will present an overview of recent experimental progress in this field, and focus in particular on the development of such a source driven by a waveform-controlled near-single-cycle terawatt laser at 1 kHz repetition rate. At the highest laser intensities ($I > 10^{18}$ W/cm^2), the plasma mirror is driven in the relativistic regime and emits UV-XUV spectra up about 30 eV photon energy (i.e., down to 40 nm wavelength). These can be made to be spectrally continuous, corresponding to intense isolated attosecond light pulses. Simultaneously and precisely synchronized with the light, MeV-kinetic-energy electron bunches are emitted through vacuum-laser acceleration, with ~10 pC charge and a wide 50° FWHM divergence. In a much smoother plasma density gradient, the XUV emission ceases, but another electron acceleration mechanism, namely laser-wakefield acceleration in the underdense part of the plasma, produces a much brighter electron beam with 25° FWHM divergence and ~50 pC charge. In another lower-intensity interaction regime and with the steepest plasma density gradient, proton beams with up to 100 pC bunch charge, 0.4 MeV cut-off energy and a divergence as low as 3° are emitted in the target normal direction.

L20

Micro- and Nanopatterning of Bionic Materials

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Micro- and Nanopatterning of Bionic Materials is a rapidly growing field to tailor special industrial, medical, and scientific applications. This is significantly driven by the exciting properties of micro- and nanopatterned materials found in natural biological species, including pronounced adhesive and anti-adhesive properties, wetting and directional fluid transport, and control of cell growth. The structuring techniques addressed here focus on developments in ultrafast laser processing. This includes direct writing techniques such as direct laser-writing and two-photon polymerization as well as the self-organized formation of micro- and nanopatterns at surfaces induced by exposure to laser radiation.

L21

High Resolution 3D Printing and Bioprinting with Femtosecond Lasers

Aleksand Ovsianikov

TU Wien

Multiphoton lithography (MPL) is set of methods offering a possibility of 3D structuring of a variety of materials at a high spatial resolution, unmatched by other additive manufacturing approaches. MPL relies on the nonlinear absorption of femtosecond laser pulses to induce photochemical processes, not necessarily limited to photopolymerization alone. An increasing portfolio of available materials enables utilization of the versatile capabilities of MPL, from producing complex volumetric 3D structures by means of cross-linking, to creating void patters within hydrogels already containing living cells. In this regard, MPL offers numerous possibilities for biomedical and tissue engineering research, as well as microfluidic and organ-on-chip applications. In this presentation, our recent progress in this area will be discussed.

L22

Nanoscale Structured Light-Matter Interactions – Unique Applications and Promising Opportunities

Peter Banzer

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Light-matter interactions are at the heart of countless modern applications. By taking advantage of light's inherent and tailorable structure, we explore new fields of application, develop advanced techniques, and study exotic phenomena. After an introduction to nanoscale light fields, we will highlight selected applications and effects.

L23

Ultrashort THz pulse generation and time resolved measurements in the THz range on solids and biological materials

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Terahertz (THz) spectroscopy and THz science in general has started about 30 years ago, when fs laser-based generation and detection of far-infrared pulses were established. These techniques make it possible to measure easily the temporal shape of the generated THz pulses, and with this perform time-domain-terahertz spectroscopy (TDTS). A TDTS measurement can result simultaneously both absorption- and index of refraction spectrum.

Generation of THz pulses having energy on the μJ level [1], made it possible to perform pump -probe type measurements on the THz spectral range [2,3]. In this way, it was possible to follow the ultrafast dynamics of electrons [3] and lattice excitations [2, 4]. High energy and high field THz pulses can be also used to control material excitations [5], and material structure [6], respectively.

Using THz radiation in another way, ultrafast charge transfer in bacteriorhodopsin was investigated [7] by observing the temporal shape of the electric field of THz radiation emitted by the biological sample illuminated by ultrashort laser pulses.

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L24

Ultrafast Laser Micro and Nanoprocessing for Biomedical Applications

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In an ageing society, medical implants are frequently used to replace damaged organs, structures and tissues in the human body. For the comfort and safety of the patient, it is essential to ensure a perfect implant integration, which implies a decent cell adhesion for tissue regeneration, and no bacteria proliferation for undesirable inflammation. Since the wide variety of microscopic living forms in contact with the implant in the body, including bacteria, soft tissue and bone forming cells, are known to be sensitive and reactive to the surface topography, it is paramount to optimize the surface of the implants for the desired integration. For dental implants, for instance, this implies reducing bacteria settlement near the prosthesis and increasing the roughness to improve implant-bone interaction and thus osseointegration. Ultrafast laser is a powerful tool for modifying the surface of medical implants, at the micro- and/or nano-scale, in order to improve or limit their cell adhesion ability. In this communication, laser micro-/nano-processing and living microorganisms' response to laser texturing are discussed. Due to its biomedical application nature, it is of key importance to ensure that laser patterning does not induce chemical or structural modifications at the implant surface. To this end, X-ray Diffraction (XRD), Scanning Transmission Electron Microscope (STEM) and Electron Energy Loss Spectroscopy (EELS) analyzes are being deployed to investigate the surface of implants after laser irradiation.

L25

Comparison of the ablation dynamics of gold immersed in air and water by pump-probe microscopy

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Laser ablation in liquid is a versatile and environmentally friendly method to generate nanoparticles from virtually any material. Since the inception of the technique in the early 1990s, a substantial research effort has been undertaken in order to bring the technique to maturity. The dynamical processes occurring during the laser ablation process of a target immersed in liquid are fundamental for understanding the underlying physical and chemical processes and nanoparticle formation mechanisms. Up to date, most of the experimental studies investigated the involved processes on timescales ranging from nanoseconds to microseconds. However, the ablation dynamics occurring on a sub-nanosecond timescale are of special importance, as the conditions under which nanoparticles are generated are established within this timeframe. Here we visualize the complete spatio-temporal picosecond laser induced ablation dynamics of gold immersed in air and water using ultrafast pump-probe microscopy. Transient reflectivity measurements reveal that the water confinement layer significantly influences the ablation dynamics on the entire investigated timescale from picoseconds to microseconds. This includes the electron injection and subsequent formation of a dense plasma on a picosecond timescale, the confinement of ablation products within hundreds of picoseconds, and the generation of a cavitation bubble on a nanosecond timescale. Moreover, we are able to locate the temporal appearance of secondary nanoparticles at about 600 ps after pulse impact. The results support computational predictions and provide valuable insight into the early-stage ablation dynamics governing laser ablation in liquids.

L26

Ultrashort-pulse laser excitation of dielectric materials: New knowledge from thin samples?

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Much information about the ultrafast laser excitation of dielectric materials has been obtained by femtosecond pump-probe experiments: an intense pump pulse excites the material by 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. This, among other things, gives rise to rapid changes in the optical properties, which can then be interrogated by a second laser pulse, the probe pulse. Previous work has supported the general excitation picture and, in some cases, even demonstrated a reasonable quantitative agreement with models of the excitation mechanisms, e.g., based on the so-called multiple-rate-equation description.

In this talk, we will present recent experimental results from our group extending ultrafast excitation to ultrathin (~1 micron thick) dielectric samples. The samples consist of Al_2O_3 grown by RF-magnetron sputtering on single-crystal silicon (100) wafers at a deposition temperature of 770 K. Subsequently, the silicon substrate is etched away in selected areas, leaving freestanding Al_2O_3 films of thickness of about 1 micron.

The purpose of this investigation is to obtain new knowledge about the excitation process by reducing the effect of laser pulse propagation through the traditionally very inhomogeneously excited dielectric samples. Preliminary investigations indicate that this technique reveals a quite different relaxation dynamics after excitation: As usual, the transmission of the sample drops significantly after excitation due to the “free” electrons in the conduction band. However, for the thin samples, we observe that the transmission seems to recover much faster than for thicker Al_2O_3 samples. We will discuss possible reasons for the observed discrepancy.

L27

Femtosecond Single-Pulse and Orthogonal Double-Pulse Laser-Induced Breakdown Spectroscopy (LIBS): Femtogram Mass Detection and Chemical Imaging with Micrometer Spatial Resolution

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Femtosecond laser-induced breakdown spectroscopy (fs-LIBS) is employed to detect tiny amounts of mass ablated from macroscopic specimens and to measure chemical images of micro-structured samples with high spatial resolution. Frequency-doubled fs-pulses (length 400 fs, wavelength 520 nm) are tightly focused with a Schwarzschild microscope objective to ablate the sample surface. The optical emission of laser-induced plasma (LIP) is collected by the objective and measured with an Echelle spectrometer equipped with an intensified CCD camera. A second fs-laser pulse (1040 nm) in orthogonal beam arrangement is reheating the LIP. The optimization of experimental setup and measurement parameters enables us to record single-pulse fs-LIBS spectra of 5 nm thin metal layers with an ablated mass per pulse of 100 fg for Cu and 370 fg for Ag films. The orthogonal double-pulse fs-LIBS enhances the recorded emission line intensities (2-3×) and improves the contrast of chemical images in comparison to single-pulse measurements. The size of ablation craters (diameters as small as 1.5 µm) is not increased by the second laser pulse. The combination of minimally invasive sampling by a tightly focused low-energy fs-pulse and of strong enhancement of plasma emission by an orthogonal high-energy fs-pulse appears promising for future LIBS chemical imaging with high spatial resolution and with high spectrochemical sensitivity.

L28

LIPSS on pre-patterned surfaces

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IESL - FORTH

Laser-induced periodic surface structures (LIPSS) or ripples are usually produced on polished surfaces with minor or random defects. Intensive experimental observations of LIPSS over a wide range of materials have been accompanied by theoretical studies. Still, it is very challenging to understand in deep in which way we can control the periodicity of these structures and how each mechanism contribute on the final morphology. Here we investigate the impact of the surface morphology on the spatial period of ripples by irradiating polished pre-patterned surfaces with a well-defined initial periodicity.

L29

The role of early mechanical surface expansion for the pulse duration dependency of the ablation efficiency in the CrMnFeCoNi high-entropy alloy

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Studies on ultrashort pulse laser-matter interaction on metal surfaces with respect to material damage or ablation have a fluence range of a few hundred mJ/cm² to a few J/cm². Hereby the observed decrease in ablation efficiency (volume removed per pulse energy) with increasing pulse duration, especially in the context of the influence of material parameters on ablation dynamics, poses a current research question. Using a heuristic ablation model, we find that the ablation efficiency is directly related to the energy deposition depth prior to ablation, called the effective penetration depth, which in turn consists of an optical, thermal, and mechanical contribution. For materials with minimal thermal conductivity and direct lattice heating, through high electron-phonon scattering rates, the aforementioned contributions to the effective penetration depth become separable. High-entropy alloys meet these material requirements and therefore the CrMnFeCoNi alloy was investigated in this work. We present experimental results on single pulse laser ablation of CrMnFeCoNi for pulse durations between 500 fs and 20 ps. The crater surface topology and ablation energetics are discussed in the context of the presented ablation model assuming an expanding surface starting within the first ps after pulse impact. We find that high-entropy alloys exhibit maximum stress confinement due to their strong electron localization. The outstanding decrease in ablation efficiency for pulse durations longer than 3 ps is attributed to the coupling of the pulse energy in an already expanding surface.

L30

Identifying malignant tissue using fs-LIBS and machine learning algorithms

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In the treatment of cancer, surgery is often the most important and effective therapeutic method. Early detection and complete elimination of cancer cells, ensure the highest long-time survival rate for patients. The surgeon must completely remove the cancerous tumor without affecting much of the neighboring healthy tissue and the establishment of oncological safety margins is most often done during the operation by a pathologist on frozen tissue sections. This analysis is time-consuming (at least 20 minutes) because the samples must be frozen, sectioned, stained, and then analyzed.

We investigate a new approach of minimizing the time of discrimination between malign and benign tissue by an in situ, non-contact spectroscopic analysis. In a proof of principle experiment, a plasma is generated by focusing an 800 nm femtosecond laser on pathologic postoperative samples and the resulting plasma radiation with the element-specific emission is recorded as a spectrum by a gated ICCD. Preliminary pathological analysis of a slide from the same sample allows accurate knowledge of the type of tissue analyzed. Advantages of using ultrashort laser pulses compared to nanosecond ones are that the bremsstrahlung, which produces a continuous spectrum, is much lower [1] and the spectral information is obtained from a very small volume [2, 3]. Since the recorded spectra are complex and very similar to each other, Machine Learning algorithms are employed to identify them with high accuracy. In this contribution, we present the results obtained in acquiring the experimental parameters that allow the best possible differentiation for mammary and hepatic tumors.

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L31

Femtosecond nano-videography of charge carrier dynamics in atomic-scale systems

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Advancements in the fields of electronics, quantum- and nanotechnology require a nanoscale understanding of the internal processes in custom-tailored materials. Since these elementary building blocks of condensed matter are in constant motion, ultrafast temporal resolution is paramount.

In this talk, I will show how femtosecond nanoscopy reveals the dynamics of photoexcited charge carriers in custom-built van der Waals (vdW) heterostructures. I will introduce a novel non-invasive terahertz nanoscopy technique to trace charge dynamics in both conducting and non-conducting materials [1]. We demonstrate ~40 nm spatial and sub-cycle temporal resolution and probe the interlayer tunneling across an atomically sharp WSe_2/WS_2 interface. Pronounced variations of the formation and annihilation of optically bright and dark excited states emerge as a result of nanoscale strain and changes in atomic registry. The results demonstrate how ultrafast nanoscopy is indispensable in the study of intrinsically disordered vdW materials.

On a WSe_2 homobilayer, we precisely tune the density of photogenerated excitons and trace the transition of a gas of strongly bound excitons into an electron-hole liquid [2]. Revealing this excitonic Mott transition on the nanoscale allows us to circumvent averaging over unavoidable nanoscale inhomogeneities and extract the true continuous nature of the process. We find variations in the transition not correlated with topography, indicating that the exciton binding energy can be modulated on length scales inaccessible to other non-contact ultrafast probes. In the future, the technique could unveil the interplay between excitons and a broad range of quantum phases in real space.

Furthermore, lightwave scanning tunneling microscopy has been shown to induce local forces, which choreograph a coherent structural motion of a single-molecule switch in its electronic ground state [3]. The molecular switch even enables quantitative sampling of atomic-scale waveforms inside the tunnelling junction [4]. I will discuss how such breakthroughs and their combination in ultrafast multi-messenger scanning probe microscopes may soon allow us to tailor (bio)chemical reactions or ultrafast phase transitions, on their intrinsic atomic-scale spatial and fs-temporal scales.

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Poster Abstracts

P01

Measurement of Four Photon Absorption Coefficient in Lithium Niobate by Z-scan Technique

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In the past two decades optical rectification of femtosecond near-infrared pulses in lithium niobate (LN) has become the most widespread way to generate high energy terahertz (THz) pulses in the low part of the THz spectrum (0.1 – 2 THz). Recently, substantial effort has been made to overcome the limitations of the conventional tilted-pulse-front pumped setup and more detailed theoretical models have been created to compare the performances of the new suggested setups.

Despite one of the main limitation factors on the maximum usable pump intensity being the free-carrier absorption in the THz range induced by multiphoton absorption of the pump, the four-photon absorption (4PA) coefficient of the LN has not been directly determined yet.

To determinate the 4PA value of LN open aperture z-scan measurements were performed. From the minimum value of the z-scan curve the 4PA coefficient can be calculated by a simple theoretical model.

The z-scan measurements were made on congruent and stochiometric LN crystal with different MgO doping concentrations. Comparing the 4PA value of these crystals can help to select the best type of LN crystal for THz generation and for other nonlinear optical phenomena, which require high pump intensities.

P02

Development and characterization of a resonant scanner based 2-photon polymerization printer

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2-Photon Polymerization (2PP) is a 3D printing method that enables the creation of features in the sub-micrometer range. While high resolution is the technology's main advantage, it also results in a significant drawback, namely the low throughput of 2PP systems. In recent years, significant advancements in this regard were made, mainly by using galvanometer scanners instead of linear translation stages for beam positioning. With such scanners, printing velocities in the range of 1 m/s are possible. Achieving even higher speeds is possible by operating one of the scanners at its resonance frequency. The continuous movement of such a resonant scanner means that it is not possible to control the position and velocity of the scan mirror directly. Therefore, when using a resonant scanner, a particular emphasis has to be put on fully characterizing the system to be able to tailor the control signals accordingly.

We built a 2PP setup optimized for the advantages of a resonant scanner and developed test routines to characterize the system. A position-sensitive detector was used to record both the resonant and the regular galvo scanners' movement patterns. Subsequently, position scaling and laser power were corrected accordingly. This enables us to print structures with a maximum scan velocity of 66 m/s while maintaining high spatial resolution.

P03

Material Characteriation of Up-Scaled 2PP Structures

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Two-Photon Polymerisation (2PP) is a versatile tool within additive manufacturing that allows to create 3-dimensional structures with complex designs showing feature sizes down to <100 nm. 2PP utilizes nonlinear absorption of femtosecond laser pulses to induce cross-linking in photo-polymers with a high temporal and spatial control. Due to the high spatial resolution the fabrication process has always been very time consuming and hence typical overall part sizes have been within the micro-scale. However, recent advances in 2PP technology have enabled the fabrication of up-scaled objects reaching the meso- and even macro-scale while maintaining feasible processing times. As a result, 2PP now bridges the unfilled gap between micro-stereolithography (µ-SLA) and nanofabrication technologies such as photolithography. Hence, reliable and standardized characterization methods for 2PP processed materials are highly demanded now. Until now the materials characterisation of 2PP printed structures was only possible on the micro and nano-scale with non-standardized methods such as nanoindentation or atomic force microscopy, that are not capable of providing a full picture of the material properties. Up-scaled 2PP processing enables to create macro-sized test-structures that can be characterized with standardized and well-established methods (e.g., tensile tests, 3-point bending tests, …). Furthermore, by down-scaling the used test procedures it is possible to also characterize meso-sized structures. With these methods, new information on a broad spectrum of material properties can be acquired allowing it to compare 2PP processed materials with other classes of materials or differently processed materials on the meso-scale.

P04

Femtosecond laser cell surgery and wound healing on Drosophila embryos

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We present our studies on processing soft biologic materials by using bandwidth-limited and temporally shaped femtosecond laser pulses. Based on our experience in processing dielectric materials on the sub-micrometer scale [1, 2] we applied similar techniques to enhance the cell-poration efficiency with a focused laser beam in liquid environment by using single temporal Airy pulses, instead of bandwidth limited pulses [3, 4]. Recently we combined fluorescence microscopy and fs-laser tissue ablation on a living Drosophila embryo to investigate wound closure. In contrast to a typical larger scale wound of a UV laser, damage can be precisely targeted on single cells or membranes with femtosecond lasers. Thousands of bandwidth-limited pulses from an 800 nm Ti:Sapphire high energy oscillator were selected with a Pockels cell and coupled into a confocal fluorescent microscope with a high NA objective. 3h old embryos were dechorionated, washed, mounted in halocarbon oil between a microscope slide and a cover slip, and imaged with a 488 nm Ar ion laser for up to 30 minutes after fs-treatment. The optimization of the ablation process is performed by precisely controlling the energy of the pulses, the exposure time as well as adjusting the temporal pulse profile of the ultrashort laser pulses. Recent experiments on Myosin-GFP-labeled embryos with ablation in different stages of development show the migration of the myosin protein towards the damage over time. In order to produce even narrower and deeper damage, we introduce temporally asymmetric fs-pulses (pulse shaper +600 kfs^3 TOD) to our setup. Preliminary experiments in soft material (gelatin) were showing a big difference between the damage made with multi-shot BWL pulses and multi-shot TOD, in the same focusing conditions.

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P05

Nanoscopic Modular Systems for the Development of Cellular Uptake Strategies

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Nanoscopic modular systems enable the precise and controlled imitation of complex biological systems. We design synthetic bioarchitectures to develop systems for medical diagnostic and therapeutic purposes. These designed structures are highly ordered arrays self-assembled from engineered molecular modules.

We specifically aim to design and produce synthetic bioarchitectures centered around the well-established bacterial/archaeal surface layer (S-layer) protein material.

We have designed molecular cages which we can use as interrogative components and which are considered highly relevant for disease treatments. To this end, we will exploit a variety of such selected biological structures, to play a pivotal role in the context of cellular uptake.

- A SARS-CoV-2 RBD and RBM motif have been fused to the S-layer protein, SbpA via genetic engineering. The resulting protein material has been purified and tested for their crystallization behavior.
- In a collaboration with Prof. Ivo Hofacker, University Vienna, we have identified translation initiation and the conformation of mRNA as the main factors governing expression efficiency of membrane proteins [1]. Based on a translation initiation model, we optimize the expression potential for our designed constructs.
- Thus, we generated protein material to probe the genuine ability of S-layer assemblies to be taken up spontaneously or via specific entry receptors. For the microscopical analysis, we have designed a S-layer – GFP variant, which we use as fluorescence marker.

We present first results regarding biocompatibility and first uptake studies into a mammalian cell line. Our data support the impression of substantial uptake of S-layer protein nano-architectures into living human cells.

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P06

A pulsed laser-based synthesis route for formation of Au/Si nanoparticles in distilled water

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Multicomponent nanostructures combining different elements are usually synthesized by some chemical methods. These nanostructures consisting of several elements reveal a large research interest being served for various aspects in the field of biomedicine [1,2]. However, using chemical-based methods for synthesis can considerably obstruct their applications in biomedical fields due to their contamination by chemical residuals. Nanocomposite structures containing both metals and semiconductors are expected to have important practical applications due to the fact that they exhibit surface plasmonic and excitonic resonance simultaneously. It is proven that laser ablation method can be efficiently employed for the synthesis of silicon-based nanoparticles (NP). As a result, they show wide prospects for applications in various fields of biomedicine, e.g., as contrast agents for magnetic resonance or optical imaging [3,4].

Laser-synthesized metallic-semiconductor nanocomposites have already demonstrated promising perspectives for molecule detection using surface-enhanced Raman scattering (SERS) or infrared absorption techniques (SEIRA) having tracking features as Raman modality or paramagnetic defect labels at the same time [5,6]. Variable experimental conditions lead to the tuning of their plasmonic, and magnetic features influenced by physico-chemical reactions during nanomaterial synthesis. However, despite intensive investigations of laser-synthesized bimetallic nanoparticles, manufacturing of nanostructures combining both metallic and semiconductor elements by means of laser ablation technique is still challenging. Nevertheless, their promising biomedical applications for simultaneous diagnostic and therapeutic purposes require a deeper study of occurred physico-chemical processes caused by ultrafast lasers and understanding of corresponding mechanisms of the interaction between semiconductor and metallic clusters or atoms.

In this work we performed the synthesis of gold-silicon nanocomposites with dual modalities by direct femtosecond laser ablation approach in deionized water and characterized by structural and optical techniques. A method of chemical content variation is developed allowing fine-tuning of ratio between gold and silicon atom values. We also studied the dynamics of the formation of both single- and multi-element Si- and Au-based NPs.

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P07

Spot size dependence of blister formation in picosecond-laser irradiation regime for blister-based LIFT

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Laser-induced forward transfer (LIFT) is a micro-scale printing technique that allows high-resolution transfer of small volumes of liquid or solid materials from a source film onto a receiver substrate. Blister-based laser-induced forward transfer (BB-LIFT) is a variant of the LIFT technique that allows soft and clean transfer of materials and is particularly attractive for delicate nanosystems and biomolecules. In this method, a thin metal film deposited on a transparent substrate is used as an intermediate absorbing layer. The film is irradiated through the substrate by a laser pulse which generates a transient mechanical deformation (blister) thus softly ejecting materials located on the film.

In this work, we investigated the effect of the irradiation spot size on the blister formation. A sample consisting of a 250 nm thick titanium film on a glass substrate was irradiated by 6 ps pulses from a Yb:KGW laser at a 1030 wavelength under single-shot conditions. The spot size on the sample was varied by changing both focusing optics and the optics position with respect to the sample. Four optical focusing systems, lenses with 400 and 35 mm focal length and objectives with 10 and 20 magnifications, were used in the experiments. By integrating a CCD camera into the set-up, we visualized the produced blisters in-situ. We have generated blisters in the size range of 2-200 μm and demonstrated that laser fluences needed to produce the blisters are size-dependent. Among the optical configurations investigated, the short 35-mm lens is found to be the optimal choice in terms of blister size range generation since it allows scaling of the blisters from 4 μm (in focal plane) to 140 μm (out of lens focus). The smallest blisters of 2 μm obtained with the 20 objective determines the LIFT printing resolution of our set-up. The blister morphology, effects of self-focusing of picosecond laser pulses in the glass substrate and ways for optimization of the BB-LIFT process will be discussed.

P08

Ultrafast hot charge carrier transport across graphene nano-gaps

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We study the hot charge carrier transport across nanoscale junctions for ultrafast electric pulse generation on the nanometer scale. To avoid laser ablation problems common to metal-based photoemission devices, we investigate the possibility of graphene nanojunctions positioned on a sapphire substrate. We create the emitter-collector structure by bisecting a graphene strip utilizing a helium ion beam to create a ~30 nm wide nano-gap. Due to substrate interaction with the helium ion beam this gap is filled by a bulge of highly defected sapphire. Measuring the ultrafast and time-integrated charge carrier transport, we find evidence of an ultrafast photoemission across this gap. Our work demonstrates that graphene-based nano-gaps have the potential of replacing photoconductive switches at low temperatures.

P09

Sub-diffractional two photon lithography for 3D thrombocyte aggregation testing

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The minimal structure sizes in three-dimensional, multiphoton polymerization lithography (MPL) can be minimized using techniques inspired by stimulated emission depletion (STED) microscopy [1-3]. Combining MPL with STED-inspired lithography allows for fast writing of coarse scaffolds, topped with 50 nm sized anchors [4] for proteins. Using different acrylate resins [5], one can achieve, that proteins adhere only on the anchors, but not on the scaffolds [6]. It is even possible to fix one sort of proteins on specific locations on a substrate, while another type of proteins is freely moving around them within a lipid bilayer [7]. On top, such structures can be written inside microfluidic channels. We show a microfluidic device enhanced with MPL structures carrying STED-lithographically written nanoanchors that promote binding of the von Willebrand factor (vWF). The density of vWF is adjusted by varying the number of the nanoanchors on a 3D scaffold. This allows us to study the impact of the density of vWF on the activation of thrombocytes [8]. The activation of the thrombocytes seems to decrease with the density of vWF on the 3D scaffolds inside the microfluidic channels.

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P10

Femtosecond Laser Induced Surface Micro-Structure Building by Material Ejection and Ablation on Cu and Al

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A novel additive surface structuring process is devised, which involves localized, intense femtosecond laser irradiation. The irradiation induces a phase explosion of the material being irradiated, and a subsequent ejection of the ablative species that are used as additive building blocks. The ejected species are deposited and accumulated in the vicinity of the ablation site. This redistribution of the material can be repeated and controlled by raster scanning and multiple pulse irradiation. The deposition and accumulation cause the formation of µm-scale three-dimensional structures that surpass the initial surface level. The above-mentioned ablation, deposition, and accumulation all together constitute the proposed additive surface structuring process. In addition, the geometry of the three-dimensional structures can be further modified, if desirable, by a subsequent subtractive ablation process. Microstructural analysis reveals a quasi-seamless conjugation between the surface where the structures grow and the structures additively grown by this method, and hence indicates the mechanic robustness of these structures. As a proof of concept, a sub-mm sized re-entrant structure and pillars are fabricated on aluminum substrate by this method. Single units as well as arrayed structures with arbitrary pattern lattice geometry are easily implemented in this additive surface structuring scheme. Engineered surface with desired functionalities can be realized by using this means, i.e., a surface with arrayed pillars being rendered with superhydrophobicity.

P11

Cell growth behavior on laser-structured screw-like medical implants

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Cell growth behavior has shown to be influenced by physical and chemical modification of surfaces. Medical implants, such as bone implants and dental implants, require a certain cell behavior to target a specific medical problem. In case of bone implants for fracture fixation, the cells should not encapsulate the implants, so that after the healing the implant can be removed easily. In case of dental implants, it is required that the medical devices have a strong connection with the jaw bone. It is therefore desired to enhance cell attachment and growth on these kinds of implants. In this work fs-laser irradiation of pre-anodized bone screws and unanodized, sand-blasted dental screws were used to achieve the desired cell behavior. It was shown that cell growth was reduced on pre-anodized, laser-structured bone screws, while enhanced cell growth was achieved on unanodized, sand-blasted and laser-structured surfaces.

P12

Femtosecond laser activation of transparent dielectrics for selective chemical deposition of copper micro-trace

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The current transparent conductive film market is dominated by expensive and fragile Indium-tin oxide electrodes, also known as ITO. However, ITO is made from the rare metal Indium. It results in a high-cost and brittle technology. Another alternative could be a metallic mesh with metal wires less than 5 µm in diameter to be invisible but still electro-conductive. Selective Surface Activation Induced by Laser (SSAIL) is a technology for circuit traces formation on various dielectric material. SSAIL is a three-step process [1,2]: The first is surface modification by a laser; the second is chemical activation of laser-modified areas, and the last step is metal deposition by electroless plating. Originally the picosecond laser is used for the first step, however Picosecond ranged pulse duration provides unwilled thermal effects, thus limiting to form very narrow structures. In this research we enabled femtosecond laser for dielectric surface activation. The result showed that the use of ultrashort pulses in the SSAIL process enabled to plate of very narrow copper lines below one micrometre in a width, beyond physical limits of laser-spot based resolutions.

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P13

Multi-photon lithography on hydrogels for organ-on-chip applications

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Multi-photon lithography (MPL) exploits the ability of femtosecond lasers to penetrate transparent materials, and trigger non-linear absorption phenomena only in the focal volume of a microscope objective. Applying its 3D patterning capabilities to hydrogels allows researchers to recreate the microenvironment surrounding cells, providing high-resolution building blocks for organ-on-chips. Cells can invade the hydrogel structure once it has been produced, or be already encapsulated into it (or both), since most MPL processes can be biocompatible if the appropriate photochemistry is chosen. Different effects can be achieved depending on the materials, such as polymerization of the hydrogel, cleaving of an already crosslinked network, or grafting additional molecules to the polymer backbone to modify the local environment. We demonstrated several applications, including semi-permeable barriers for signals, nutrients, and drugs exchange, microchannel networks for cell invasion and microvessel formation, and guided cell migration.

P14

Fluence dependence of the laser fragmentation of gold nanoparticles in liquids: Molecular dynamics study

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Short pulse laser irradiation of a colloidal solution of nanoparticles is an effective method producing a population of nanoparticles and atomic clusters with desired properties. To investigate the mechanisms involved in the fragmentation, we develop a computational model capable of realistic treatment of a variety of interrelated processes occurring on different time and length scales, from the electronic excitation by the laser pulse, to the electron-phonon energy transfer and an explosive phase decomposition of the superheated nanoparticle, and to the generation and collapse of a nanobubble in the liquid environment. The application of the model to simulation of laser fragmentation of a Au nanoparticle in water has revealed two distinct formation channels of fragmentation products. The first channel involves the direct injection of compact nanodroplets propelled by the phase explosion of the irradiated nanoparticle deep into the water environment. The second formation channel involves a more gradual growth through agglomeration of numerous atomic clusters embedded into a narrow water region surrounding the laser-induced nanobubble. This channel can produce irregularly shaped nanoparticles and leads to a rapid decline of the population of atomic clusters on the timescale of nanoseconds. All the clusters and nanoparticles experience an ultrafast quenching by the water environment. They feature a high density of twin boundaries and other crystal defects, which can serve as chemically active sites for catalytic applications.

The extension of the computational study to analysis of the dependence of the fragmentation process on the energy density deposited by the laser pulse has revealed several distinct regimes of the nanoparticle fragmentation. The decrease of the energy density from 80% of the energy required to fully vaporize the Au nanoparticle down to 60% and 40% of the vaporization energy is found to lead to qualitative changes in the fragmentation mechanism. In contrast to the explosive fragmentation and prompt injection of the fragmentation products into the water environment surrounding the nanobubble observed at the high energy density, a big fraction of the nanoparticle material remains inside the nanobubble at lower energy densities, and the fragmentation produces a large central fragment surrounded by smaller satellite fragments. The origin and size of the central and satellite fragments are rather dissimilar at the two lower energy densities, which reflect the existence of two distinct low-energy fragmentation mechanisms revealed in the simulations. The computational predictions have important practical implications for achieving an improved control over the size, shape and defect structures of nanoparticles produced by laser fragmentation in liquids.

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Enhancement of Spectral Broadening in Fused Silica Using Femtosecond Vortex Laser Pulses

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The spectral broadening of ultrashort laser pulses plays an essential role in nonlinear laser physics [1], optical frequency metrology [2] or pulse compression in time domain [3]. Wide bandgap materials (sapphire) or transparent dielectrics (fused silica) are often used for supercontinuum generation, most often in the form of photonic structures, e.g., hollow-core fibers.

In this work we focus on spectrum broadening in fused silica plate of near-IR ultrashort laser pulses with different pulse shapes and polarization states. We show that in the case of donut-shaped laser pulses, spectral broadening is more effective for optical vortices than for radially or azimuthally polarized beams with a zero-orbital momentum. Similarly, we observe a wider spectrum for circular polarization than for linear for Gaussian beams.

Linear polarizations also result in higher nonlinear absorption, which is undesirable for supercontinuum generation. The decrease in nonlinear absorption for circular polarization is also evident from the observation of luminescence of self-trapped excitons in a strongly nonlinear multiphoton regime. We observe that the charge carrier generation rate drops significantly in the case of circularly polarized light which is the consequence of intra-cycle carrier dynamics [4].

[1] J. Piel, M. Beutter, and E. Riedle, Optics Letters 25, pp. 180-182 (2000).
[2] Th. Udem, R. Holzwarth, and T. W. Hänsch, Nature 416, pp. 233–237 (2002).
[3] B. Schenkel, R. Paschotta, and U. Keller, J. Opt. Soc. Am. B 22, pp. 687-693 (2005).
[4] M. Kozák, T. Otobe, M. Zukerstein, F. Trojánek, and P. Malý, Phys. Rev. B 99, pp. 104305-10414 (2019).

	Monday, March 14	**Tuesday, March 15**	**Wednesday, March 16**
8^{30} - 8^{45}	**Registration**		
8^{45} - 9^{00}			
9^{00} - 9^{15}	**Opening and greetings**	Lecture 10 **Stoian**	Lecture 22 **Banzer**
9^{15} - 9^{30}	Lecture 1 **Zhigilei**	Lecture 11 **Römer**	Lecture 23 **Hebling**
9^{30} - 9^{45}			
9^{45} - 10^{00}	Lecture 2 **Bulgakova**	Lecture 12 **Mauclair**	Lecture 24 **Sedao**
10^{00} - 10^{15}			
10^{15} - 10^{30}	**Posters:** **Short lectures and session** with coffee	Coffee	Coffee
10^{30} - 10^{45}			
10^{45} - 11^{00}		Lecture 13 **Bulgakov**	Lecture 25 **Spellauge**
11^{00} - 11^{15}			
11^{15} - 11^{30}		Lecture 14 **Sanner**	Lecture 26 **Balling**
11^{30} - 11^{45}			
11^{45} - 12^{00}	Lecture 3 **Rethfeld**	Lecture 15 **Horstmann**	Lecture 27 **Giannakaris**
12^{00} - 12^{15}			
	Free discussion	Free discussion	Free discussion
17^{00} - 17^{15}	Coffee	Coffee	Coffee
17^{15} - 17^{30}	Lecture 4 **Pfeiffer**	Lecture 16 **Stratakis**	Lecture 28 **Maragkaki**
17^{30} - 17^{45}			
17^{45} - 18^{00}	Lecture 5 **Derrien**	Lecture 17 **Courvoisier**	Lecture 29 **Redka**
18^{00} - 18^{15}			
18^{15} - 18^{30}	Lecture 6 **Biegert**	Lecture 18 **Hrabovsky**	Lecture 30 **Sarpe**
18^{30} - 18^{45}			
18^{45} - 19^{00}	Coffee	Coffee	Coffee
19^{00} - 19^{15}	Lecture 7 **Hallum**	Lecture 19 **Haessler**	Lecture 31 **Huber**
19^{15} - 19^{30}			
19^{30} - 19^{45}	Lecture 8 **Aeschlimann**	Lecture 20 **Heitz**	**Closing remarks**
19^{45} - 20^{00}			
20^{00} - 20^{15}	Lecture 9 **Kautek**	Lecture 21 **Ovsianikov**	
20^{15} - 20^{30}			