Book of Abstracts

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International Conference on Ultrafast Optical Science (UltrafastLight-2018), is the broad-scope, annual international symposium dedicated to the most important aspects of ultrafast phenomena in different fields of natural sciences and engineering.

The Conference topics:

1. Radiation and nuclear photonics at high fields
2. Ultrafast phenomena in condensed matter and ionized gases
3. Ultrafast laser nanofabrication and nanophotonics
4. Femtosecond non-linear optics. Filamentation.
High field THz generation.
5. Femtosecond laser photobiology and photochemistry.
6. Physics and technology of ultrashort laser pulses and innovative femtosecond laser technology.
7. Femtosecond radiation in spectroscopy and optical frequency metrology.

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Scope
Laser plasma sources of ionizing radiation
Nuclear physics with lasers
Extreme fields physics
Ultra high intensity facilities

Efficient generation of attopulses at the interaction of intense laser radiation with the shaped targets

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Recently we have proposed an efficient scheme of generation of short dense electron bunches during the interaction at large angles of incidence of a laser pulse with a limited foil target [1]. It has been shown that, upon reflection from the target, the relativistic intensity laser pulse is efficiently converted into a sequence of atto-pulses. Later [2], the generation of high-intensity atto-pulses has been investigated also in cylindrical geometry by using 3D PIC code. Due to the rotation-symmetric target, a circularly polarized laser pulse was considered, propagating on the axis of a hollow cone-like target. The large incidence angle and constant ponderomotive pressure lead to nano-bunching of relativistic electrons responsible for the laser-driven emission. We found the source and direction of the coherent radiation that ensures the existence of atto-pulses. The intensity modulation in the harmonic spectrum was well described by the extended model of coherent synchrotron emission. The spatial distribution of the higher harmonics resembles a spiral shape which gets focused into a small volume behind the target.

Since the laser pulse reflected from plasma inherently contains low-order harmonics, a second reflection from a fresh plasma surface leads to the increase of spectral intensity. In the work [3], we have made an extensive study of multiple reflections of a short pulse between two solid density plasma walls at oblique incidence, with the help of 2D PIC simulations. A loosely focused pulse is considered that can propagate between two foils over consecutive reflections without significant loss of intensity. It is shown that even in the weakly relativistic regime the intensity of high harmonics can be amplified by three orders of magnitude with help of this method thus we can increase above considered generated atto-pulses.

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High energy electrons in relativistic laser-plasma interaction

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Secondary sources of high energy particles and hard radiation, produced by the action of short intense laser pulses are widely used for creation and diagnostics of extreme states of matter. Various mechanisms of heating and generation of hot electrons in the relativistic laser interaction with solid and low-density targets are considered. In particular, an effective generation of highly energetic electrons of tens of MeV energies in near critical plasma layers is demonstrated. These collimated high energy electron beams carrying the charge that many orders of magnitude exceed the value predicted by the ponderomotive Wilks scaling produce short bursts of hard X-rays. Generation of high energy density states by the ultrahigh contrast femtosecond laser pulses interacting with a near solid step-like electron density profile is analyzed.

Experimental data on generation of hot electrons and hard radiation obtained at the facilities of PHELIX (Darmstadt) and JETI40 (Jena) are discussed.

Optimizing the dephasing length limitation in a laser plasma accelerator

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Despite the advanced techniques for improving electron acceleration conditions, the small amount of dephasing length continues to play a very destructive role in accelerating electrons. We represent that by injection of different electron profiles and beam loading effect it is possible to change the ratio of accelerating to decelerating field of the wake-field using numerical simulations. Therefore, the effective field on the accelerated electrons in the accelerating phase of the field becomes very small in decelerating phase. Following this, the electron beam loses a very small amount of energy even when it enters the decelerating phase. So, the limitation of the dephasing length is greatly reduced.

QED cascade with multipetawatt-class lasers: a road to attosecond-scale highly directed GeV gamma-ray sources

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We consider the properties of gamma radiation generated during the development of a quantum electrodynamic cascade in the field of counter-propagating multipetawatt laser beams configured in the form of a dipole wave. Principal attention is paid to the linear regime of cascade development, when the emerging plasma almost does not affect the electromagnetic field but can already be a sufficiently effective source of gamma radiation with unique properties. It is shown that in this regime the considered field configuration possessing the axial symmetry allows the generation of gamma radiation with narrow directivity (about 1 mrad for 10 PW level). Moreover, the special features of particle motion in the fields of tightly focused multipetawatt level laser beams enable generation of trains or isolated pulses of gamma radiation in the GeV energy range with attosecond-scale duration. The influence of the cascade on the spectrum of the generated radiation is revealed and the dependence of the spectrum on the total laser power is obtained.

Stochastic electron heating in combined field of several overlapping laser pulses of a picosecond duration

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Efficient electron acceleration and heating is demonstrated in a multimode structure created by overlapping of several laser beams of relativistic intensity and picosecond duration near a sharp target-vacuum interface. The process considered includes a combination of a slow stochastic heating and a fast regular acceleration in a resonance interaction with one of the wave packets. It results in formation of a population of energetic electrons with an exponential distribution in energy characterized by a high effective temperature and a sharp energy cutoff. Both these parameters depend on the number of crossing laser beams and the angles of their intersections. This process of laser-electrons interaction provides an example of efficient electron heating in vacuum electromagnetic fields without participation of electrostatic plasma waves. It might contribute to generation of a suprathermal particle population with an effective temperature significantly exceeding the commonly accepted a standard ponderomotive scaling. This work was partially supported by RFBR grants № 16-02-00088a, 18-02-00452, 17-02-00366a.

THz and gamma-ray generation from laser-plasma interaction

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The sources of secondary electromagnetic radiation based on laser-triggered electrons have broad potential applications such as medical and biological imaging, diagnostics for materials science, probing of dense plasmas, security (inspection) systems. Most of these applications require a brightness of electromagnetic radiation pulse generated during one shot that has to be enhanced. This can be achieved by choosing a proper (optimized) scheme of laser-plasma interaction. In this talk we discuss several mechanisms of THz generation in the interaction of short laser pulses with solid targets. We show how the strong THz surface wave can be triggered by light pulse at the metal wire surface and how it propagates along wire in the form of a weakly damped Sommerfeld wave. The electron bunch guiding along a wire by strong Sommerfeld surface wave field has been modeled. Such guiding is compared with experimental results.

Based on multidimensional PIC simulations of short laser pulse interaction with homogeneous planar target, we also present results of an optimization study to find the best design parameters, which maximize the number of high-energy electrons generated by subpetawatt class laser system for deep gamma radiography purpose. We demonstrate that a low-density target with electron density of one tenth of critical density irradiated by femtosecond laser pulses is optimal for acceleration of maximum number of energetic electrons suitable for the MeV bremsstrahlung gamma-ray source for deep gamma-radiography.

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Laser-triggered charged particles acceleration for nuclear and gamma sources

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Laser-driven particle acceleration by femtosecond high-power pulses is a topic of extraordinary interest for fundamental research and nuclear applications. These issues motivated a worldwide search for different mechanisms of electron and ion acceleration with the aim to maximize both the yield and the energy of the generated particles. In this context, an important role is played by low-density targets with an electron density close to the relativistic critical density that is discussed here on the basis of the 3D particle-in-cell (PIC) simulations and theoretical models.

We have extended recently published results of so-called SASL (synchronized acceleration by slow light) simulationsto other schemes of laser-plasma interaction for proton acceleration involving manipulation of laser polarization and low-density targets which are available in practice. In all cases, the main ideais to capture the protons from a target front side in laser pulse ponderomotive electric field sheath and keep them synchronized with the latter due to specific nonlinear propagation and laser-target design. We have performed optimization study for given laser pulse energy to find the target which may produce protons with maximum possible energy.

Microstructured targets in the form of planar foils with rods and sheets of sub-micron thickness and micron lengths at their front side in normal direction have also been tested in the 3D PIC simulations as neutron sources. This microstructured coating with proper chosen geometrical sizes of microstructures allows laser light to penetrate rather deep in a target to heat it volumetrically. An average density of such coating can be as high as one tenth of solid density. All this allows maximize a neutron yield.

The 3D PIC simulations have also demonstrated effective acceleration of electrons from low-density targets in terms of the increased electron yield. The electron charge per shot with energies in excess of 100 MeV reaches multi-nC level for current femtosecond lasers, that is not available for LWFA-based accelerators with a standard gas density and electron acceleration from planar solid targets with step-like density. This optimized electron source has been used to calculate neutron yields from some (gamma, n) nuclear reactions.

Our findings constitute an important approach to laser-based nuclear sources for deep gamma-radiography of dense samples, isotope production and neutron generation. This approach is demonstrated on several examples.

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Fine-foam experimental studies with long and short powerful laser pulses to present variable x-ray and particle plasma sources

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Application of low density targets are very promising in varied fields such as hydrodynamic studies, enhancement of laser driven shock pressure, smoothening of laser spatial profile to mitigate instabilities and enhancement of x-ray conversion efficiency, etc. Foam layers are being used in inertial fusion targets to improve implosion symmetry. Utilization of low-density porous materials in various types of target design is believed to be the promising approach in a number of important research programs. In the paper, we will report the experiments done on Interaction of long pulse (30 J / 500 ps) and short pulse laser system (1 J /25 fs) with low density foam targets of various kinds of materials from low Z (polymeric foam), $(CD_2)_n$, to high Z (Gold and Bismuth) having various density (2 mg/cc, 4 mg/cc, 7 mg/cc, 10 mg/cc and 20 mg/cc) In these experiments various characteristics such as transmittance to laser radiation, x-ray and ion emission studies, x-ray pulse duration, absorption processes and hydrodynamic studies were done using x-ray detectors, x-ray streak camera, Ion collectors, Thomson Parabola and multi-frame optical shadowgraphy and optical streak camera. It is observed that the laser pulse duration was shortened from 680 ps to 360 ps after passage through the targets for areal density of about 600-900 mg/cm^{-2} and laser absorption increased to 84 % in case of 2 mg/cc TAC foam target compared to 60 % for solid polymeric target.

The pulsed laser irradiation of a subcritical density target was found to give rise to harder X-rays than the irradiation of a supercritical density target. In case of hard x-rays i.e., 4 - 16 keV and 5 - 8 keV) regime it was 2.3 times higher in 2 mg/cc foam target compared to 20 mg/cc foam target. This may be due to supersonic bulk heating of the subcritical density targets by a thermal wave as well as to the absence of significant energy loss for hydrodynamic ion motion. An analysis of the ion emission revealed an increase in ion flux and a shift of ion energy spectrum towards higher energies in going to supercritical density targets.

In case of interaction of Low density TAC foam targets with the ultrashort laser system, the x-ray emission in soft x-ray (1-17 keV) and hard x-rays (>500 keV) were measured using XRD in integration mode and NaI detector by keeping XRD inside the chamber and NaI detector outside the chamber in front of 15 mm thick glass window. It is observed that the hard x-ray yield (>500 keV) is about 90 – 100 times compared to solid polyethylene targets. Thomson parabola measurements are being analyzed and will be presented in the conference meeting.

The acceleration of deuterium ions under the influence of long pulse and short pulse laser system on the $(CD_2)_n$ with densities 0.275, 0.35 and 0.44 gm/cc have been done. In case of long pulse laser system, the peak velocity is also compared for different densities of targets and it is found that peak velocity is high towards target normal for 0.44 gm/cc CD_2 foam, while at large angles, the effect of target density is small. It indicates the emission of more energetic ions from higher density CD_2 foam towards target normal in comparison to low density foams. Analysis of ion emission from ultrashort pulse is underway.

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Particle trajectory analysis of QED pair plasma interaction with petawatt level lasers

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Dense QED electron-positron plasmas presumably may be produced by the upcoming petawatt level laser facilities. As shown by modeling, at optimal focusing of such powerful radiation extreme states of a pair plasma and gamma photons in terms of densities, energies, fluxes can be reached. At higher laser powers, in the so-called pinch regime of the interaction, unprecedented magnetic and electric fields can also be generated when self-generated magnetic field compresses an electron-positron plasma column. Qualitative changes in the interaction dynamics occur when the current through the central plane z=0 exceeds approximately 20 MA. In this work, an analysis of the interaction dynamics by studying the individual particle trajectories was carried out using extensive PIC simulation. Analyzing the properties of the single particle motion, we can better understand the dynamics of the plasma. In the linear interaction mode, as well as in a nonlinear mode with a laser power not so high (< 20 PW), particle motion is determined by the ART trajectories when particles approach the maximum of the electric field. As the power is increased, the particle trajectories drastically change, since the particles are pushed to the center by means of self-induced magnetic fields forming the oscillating curved trajectories. Simulations show that the size of the compressed plasma column is ultimately limited by the size of these trajectories, which is determined by the field distribution and thus can be no less than 1–2 grid cells. By varying the grid resolution, it is possible to estimate the maximum possible compression of the plasma column and the corresponding level of emerging magnetic and electric fields.

Longitudinal field generation and ion acceleration in undercritical plasmas enhanced by radiation friction

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We review our recent 1D and 2D PIC simulations and analytical model [1,2] of propagation of an ultra-strong laser pulse in underdense plasma. We identify the parameters, for which generation of longitudinal field is substantially enhanced by radiation friction, and explain the reason of this enhancement. Furthermore, if the ions are mobile, they are rapidly accelerated by the resulting longitudinal field. We discuss the parameters of the resulting ion bunches and the impact of radiation friction, plasma thickness, and laser focusing on ion acceleration in such a scheme.

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X-ray spectroscopy evidence of plasma shell formation in experiments modeling accretion columns of young stars

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This paper aims the studying the accretion dynamics of young stars in laboratory. In order to investigate the accretion dynamics in the laboratory we create a collimated narrow (1 mm diameter) plasma stream by imposing a strong (B_z=20 T) external and uniform poloidal magnetic field onto an expanding high-power laser (1 ns duration, 10^{13} W/cm^2) ablated plasma. The stream (v_{stream}=750 km/s) propagates parallel to the lines of the large-scale external magnetic field, as in the present picture of mass accretion in Classical T Tauri stars, onto an obstacle mimicking the high-density region of the star chromosphere. We observe in these experiments that matter, upon impact, is laterally ejected from the solid surface, then refocused by the magnetic field toward the incoming stream. Such ejected matter forms a plasma shell that envelops the shocked core. The parameters of both plasma core and shell were studied by means of spatially resolved X-ray spectroscopy and Mach-Zehnder interferometry. The issue connected with current discrepancies in astrophysics between mass accretion rates derived from X-ray and optical observations have been studied analysing question of opacity in experiment through shell plasma.

Electron bunch formation under action of relativistic laser pulse onto long-scale undercritical plasma

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The generation of the electron bunches with high energy and charge in the laser-plasma interactions can occur due to various mechanisms of laser acceleration. In this paper we present experimental research of the possibility of electron bunch generation at an intensity around 10^{18} W/cm^2 in a long-scale undercritical plasma, formed by additional nanosecond laser pulse with controlled parameters.

Characteristics of electrons generated in the interaction of a laser pulse of relativistic intensity and solid targets from W and a film were experimentally investigated. We used additional nanosecond laser pulse with controlled parameters to create undecritical plasma of various scales. It has been demonstrated experimentally that electrons can be generated in the form of a beam with a divergence of ~ 0.1 rad. A technique for direct measurement of the energy spectrum of electron beams using a magnetic dispersion system has been developed and tested. Using it we can now obtain spectra in each individual shot. It is shown that for experiments with a lower particle flux for direct detection of electronic spectra, the Medipix detector can be used effectively. Electron temperatures in all the investigated cases are in the range T = 1.1 ÷ 1.8 MeV. We also changed the angle of incidence of laser radiation to the target. It was found that for the W target with an increase in the angle of incidence of radiation, the electron beam shifts to the normal. For film targets, electrons are always generated at an angle close to 90° from the direction of the original beam. We also investigated spatial correlations between electron beam and optical harmonics yield. The experimental results are supported by 2D3V PIC modeling and are in good agreement with it.

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Optical diagnostics of femtosecond laser plasmas

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The plasma formed by a powerful femtosecond laser pulse on a solid target surface has been an object of intense investigation in recent decades. Electron acceleration in such plasma is one of the research directions in this field. Studies show that plasma created by the pre-heating laser pulse plays a decisive role in such a process [1], magnetic fields generation during laser-plasma interaction can also have a significant effect [2-3].

In this research formation of plasma plume produced by a powerful nanosecond laser pulse on a solid target surface is studied experimentally, as well as interaction this pre-plasma layer with intense femtosecond laser pulse. Self-generated magnetic fields were measured with a three-channel polaro-interferometer [4]. The dynamics of the electron plasma density was also investigated by interferometric diagnostics.

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Optimization studies of high energy electron generation from nanostructure targets

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Surface and volumetrically structured targets as an alternative to traditional solid or gas targets are of great interest for laser triggered accelerations. They are often used to increase the efficiency of laser energy transfer to charged particles. Nanograss (or nanowire) and nanosheet substrates are of the most promising structures. Electron acceleration from such targets irradiated by shot relativistic laser pulse has been studied with 2D and 3D PIC simulations. We have modeled these structures as a number of cylinders (or sheets) located on an irradiated flat target surface. An Influence of various parameters of the nano structures on electron acceleration was evaluated and optimized, including the average electron density, the structure length and thickness. The have performed calculations which showed that average and maximum electron energies can be significantly increased as compared to standard flat targets. Also, the characteristic features of high-energy electrons generated in these structures have been revealed.

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Quantum effects in extreme fields - ultrahigh intensity physics with ultrafast lasers

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At the Center for Relativistic Laser Science (CoReLS) we have recently commissioned a 4 PW laser system that can focus in various geometries, reaching peak intensities of up to 10^{23} W/cm^2. We are establishing a broad scientific program centering on exploring quantum effects at ultrahigh intensities, the development of laser particle and photon sources and the development of the required ultrahigh intensity laser technology.

I will review the status of the program, describing the laser and experimental system using examples of recent and currently planned experiments on radiation reactions, electron and ion acceleration and non-linear Compton scattering and outline future plans and opportunities for collaborations and joined experiments.

Optimization of laser-plasma coupling at relativistic femtosecond interaction with solids for enhanced hot particles and high energy radiation production

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The ultrashort relativistic laser-driven hot plasma is a promising source of fast particles, X-rays and gamma radiation for tasks of nuclear reactions excitation, phase contrast imaging radiography etc. High brightness, stability at high repetition rate of laser pulses of the source in different energy ranges is a key goal for successful use of plasma source in application.

In this work we present the recent results on fast electrons acceleration and hard X-ray generation at relativistic femtosecond interaction with spatially non-uniform hot dense plasma onto the surface of solid (flat and modified on wavelength scale) targets. The plasma was formed by a laser pulse delivered by the Ti:Sa laser system (pulse duration – 50 fs, energy on target – up to 100 mJ, wavelength – 800 nm, repetition rate – 10 Hz, contrast on ps timescale $>10^9$). An artificial prepulse of nanosecond duration could be introduced (a 100 mJ pulse form Nd:YAG laser) to form a controlled preplasma onto the surface of the target. Apart from the simple flat target different modified targets prepared by laser ablation, electrochemical etching were used in the experiment.

It is found, that the efficiency of laser to plasma coupling varies appreciably on the parameters of the target to the moment of the main femtosecond pulse arrival. Substantially increased hot electrons energy (from ∼200 up to 1 MeV) was detected both, when the preplasma with specific spatial scalelength is formed by a prepulse or when the modified targets are used at high contract. High energy photons production increased one order of magnitude at these conditions. With the support of PIC simulations we revealed that in both cases the hot electron generation mechanism is complicated and involves several acceleration stage, such as plasma wave formation and breaking with subsequent energy gain in crossed laser fields in the preplasma, vacuum heating in the sub-wavelength structures at the plasma-vacuum interface.

We have also demonstrated, that the plasma source may be utilized for X-ray and gamma imaging of dense objects. With the increased brightness the necessary exposure time becomes reasonably limited for fast inspection.

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Dense e+e- plasma generation in extreme laser fields

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Recent progress in laser technology has led to designing the next generation of laser facilities, such as ELI and XCELS producing extreme electromagnetic fields. Self-sustained quantum electrodynamic (QED) cascades can be developed in such fields. In this talk we will discuss possible ways of producing electron-positron-pair plasmas of extreme densities, as well as the physical mechanisms underlying them. Using full 3D QED-PIC modeling we will also demonstrate that the time scale for the formation of extreme plasma states can be controlled by a proper choice of target-seed and laser parameters.

Experimental observation of intense magnetic field in a snail-shaped targets irradiated by a relativistic picosecond laser pulse

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Most of the known generators of quasistationary magnetic fields, realized both with and without lasers, operate due to the strong discharge current flowing through the coil. Vacuum magnetic field generated using this method can be used to guide particles or to magnetize secondary objects, for example plasma, which is relevant for laboratory astrophysical studies. A more natural and simple approach is to obtain already magnetized plasma that can retain a strong magnetic field for a hydrodynamic time scale. In the research work, carried out at PHELIX laser facility (GSI, Germany), a study of Snail-shaped targets, irradiated by a short (0.5 ps) laser pulse (50 J) was conducted. Laser pulse simultaneously produces magnetic field due to the current of accelerated electrons and discharge currents, and ablative plasma. Plasma is born in the magnetic field and therefore is magnetized. Even after the end of laser pulse the magnetic field, "frozen" in the ablated plasma, remains. The proton deflectometry used in the experimental study confirmed the presence of previously theoretically predicted [1] magnetic field with an inductance of the order of several hundred Tesla with a lifetime of more than a hundred picoseconds inside the target. The proposed setup can be used for a number of different applications from laboratory astrophysics to particle acceleration schemes.

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Magnetized plasma structures production by intense laser radiation

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Magnetized plasma is a common object in the universe and in human-produced devices. It possesses a specific internal parameter, known as magnetization, which controls a lot of physical processes both on hydrodynamic and kinetic scales. Laboratory studies, involving laser facilities, may operate with the magnetized plasmas in a wide range of energy density, up to the quantities related to astrophysical values, but generation of such plasmas appears to be a challenging problem. In this work, we present some recent results showing possibilities to obtain highly magnetized hot plasma structures, existing on the time scale more than tens of picoseconds. This is already enough to perform various experimental studies, related both to astrophysics and for perspective technologies.

Scalings of sheath-acceleration of protons driven by ultra-intense subpicosecond laser pulses

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Recently it has been experimentally shown that target normal sheath acceleration of protons from ultra-thin targets irradiated by sub-picosecond laser pulses of intensities above 10^{21} W/cm^2 is suppressed compared to well-established models [1]. This has been attributed to gigagauss-level magnetic fields generated in the target. Here we would like to present a numerical and semi-analytical investigation of the observed effect for wide range of laser pulse parameters.

Based on 2D full-scale numerical simulations performed by PICADOR code implementing a Particle-In-Cell method [2], it has been shown that at intensity of the order of 10^{21}–10^{22} W/cm^2 a scaling of a cutoff energy of the accelerated protons with intensity deviates from a well-established $\epsilon_c \sim I^{0.5}$ substantiated by a Mora model [3] for laser pulses with duration exceeding 100 fs.

This deviation is showed to be connected to effective magnetization of the hottest electrons producing at the maximum of laser pulse intensity which in turn results in lower cutoff energy of accelerated protons. The effect starts to play role when the Larmour radius of the electrons become comparable to the sheath thickness at the moment of the laser pulse maximum arrival.

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Electron acceleration and gamma-ray emission at intense laser-solid interaction

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An interaction of a relativistically intense linearly polarized laser pulse with an overdense plasma is analyzed. In low intensity regime when a laser pulse is grazingly incident onto a planar solid-state target, a slightly superluminal field structure is formed near the target surface due to the incident and reflected waves superposition. This field structure can both extract the electrons from the target and accelerate them. It is shown that the acceleration is possible and stable for a wide range of electron initial conditions. The high-energy electrons are the source of gamma-emission. The effect of a preplasma is also discussed.

In high intensity regime a dense electron layer is formed on the plasma edge which relativistic motion results in high harmonic generation, ion acceleration, and incoherent synchrotron emission of gamma-photons. A self-consistent analytical model, that describes the edge motion and apply it to the problem of incoherent synchrotron emission by ultrarelativistic plasma electrons, are presented. The plasma density and the angle of incidence of p-polarised laser pulses that correspond to the highest gamma-ray generation efficiency and high gamma-ray directivity are found. The analytical results are in agreement with 3D particle-in-cell simulations.

We have carried out numerical simulations of oblique incidence of a laser pulse with an intensity of I = 1.33×10^{23} W/cm^2 on a planar plasma layer and found the plasma density and the angle of incidence of p-polarised laser pulses that correspond to the highest gamma-ray generation efficiency and high gamma-ray directivity. The shape of the plasma surface has been determined by simulation and conditions have been considered that lead to an increase in generation efficiency.

This paper examines the effect of a density gradient produced in a target by a prepulse on the efficiency of synchrotron gamma-ray generation by a laser pulse with an intensity of the order of 10^{22} to 10^{23} W/cm^2 obliquely incident on a planar target. We demonstrate that the gamma-ray and hard X-ray generation efficiency and the fraction of absorbed laser pulse energy can exceed those in the case of a uniform target with a sharp boundary by a factor of 1.3 – 2, depending on the angle of incidence. At a moderately large preplasma size, the optimal angle of incidence is 30°. At a sufficiently large size, normal incidence is optimal.

When a relativistically intense p-polarized laser pulse is grazingly incident onto a planar solid-state target, a slightly superluminal field structure is formed near the target surface due to the incident and reflected waves superposition. This field structure can both extract the electrons from the target and accelerate them. It is theoretically shown that the acceleration is possible and stable for a wide range of electron initial conditions. Particle-in-cell simulations confirm that this mechanism can actually take place for realistic parameters. As a result, the electron bunches with a charge of tens of nC and GeV-level energy can be produced using a laser intensity 10^{21} -– 10^{22} W/cm^2. It is also shown that the presence of a preplasma can improve acceleration, which becomes possible because of more efficient electron injection into the accelerating field structure.

Ultra-high intensity laser interaction experiments at the University of Michigan

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I will review some of the recent results from experimental research into high intensity laser plasma interactions at the University of Michigan. This includes the development of "table-top" sources of electrons, x-rays and gamma rays as well as their use for probing and imaging matter with extreme temporal resolution. I will also discuss recent measurements of the dynamics of ultra-strong magnetic fields generated by relativistic laser pulses.

Intense terahertz and infrared radiation from laser pulse interaction with mass-limited and gas targets

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Generation of intense terahertz and infrared radiation with a high-intensity laser pulse is studied for interaction with mass-limited and gas targets. In the first case, fast and strongly non-stationary processes are initiated, which are characterized by considerable charge separation. A laser pulse of relativistic intensity incident normally at a mass-limited target can induce a simultaneous motion of all electrons from the target in the longitudinal direction (along the laser beam axis) due to the Lorentz force. This force accelerates electrons to relativistic velocities. As a result, a relativistic electron bunch can be formed and its electron density can be about solid target density. For relativistic electron bunch formation, the laser pulse amplitude should be larger than some threshold, which is determined by the material and geometry of the target. During interaction, ions of the target cannot move considerably from their initial position because of high inertia. Due to strong Coulomb field, after some time, a part of relativistic electrons turns back and returns to their initial position. Because of gained velocity, they oscillate around the ions. In this case, radiated pulse can contain several cycles of oscillations, its frequency depends on the system parameters and can belong to the terahertz or infrared bands. It is shown that the temporal profile of radiation depends on the laser pulse amplitude and duration, electron density of the target and its geometry. The amplitude of the radiation can be at the relativistic level for modern petawatt laser systems. Several analytical models are proposed for this process and studied.

In the second case, relatively long pulses of terahertz or infrared bands can be generated with a wakefield formed by a laser pulse in a gas medium. For increasing the interaction, a density modulated plasma can be used. Characteristics of radiation generated during this process such as polarization, frequency, amplitude, etc. are studied. Here also, analytical model is considered for generation of terahertz or infrared radiation.

Revealing the parameters of laser-plasma and FEL X-ray sources using LiF detector

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Recent achievements in characterization and control of various free electron laser (FEL) sources, as well as the laser plasma sources and the calibration data for LiF detectors for particular photon energy range, are given. Our experiments proved spatial resolution to be of $\sim$ 700 nm for soft X-ray range, and of $\sim$ 1.0 μm for 10 keV X-ray beam. Theoretical estimates on photoelectron cloud formation revealed fundamental limit on spatial resolution capability of LiF detector. High sensitivity and uniquely large dynamic range exceeding 10^6 of LiF crystal detector allowed measurements on the intensity distribution of hard X-ray SACLA XFEL beam at distances as far from the focal plane, as near the best focus. Also, considering the diffraction patterns from test objects , spectral and coherent properties of X-ray beams were revealed for laser-plasma sources.

Manipulating relativistic electrons with intense lasers: towards compact plasma accelerators

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Laser Plasma Accelerators (LPA) rely on the electrons motion control with intense laser pulses [1]. The manipulation of such relativistic electrons with lasers allows a fine mapping of the longitudinal and radial components of giant electric fields with values that can exceed hundreds of GV/m [2]. This control is crucial to optimize laser plasma accelerators for delivering ultra-short and ultra-bright energetic particle or radiation beams.

To illustrate the beauty of laser plasma accelerators I will show some concepts that give improvements the quality of the electron beam, its stability [3] and its energy gain [4], and its divergence [5].

I'll then show how by controlling the quiver motion of relativistic electrons intense and bright X-rays beam are produced in a compact and elegant way [6-8]. Finally I'll show some examples of applications.

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X-ray diagnostics of warm dense matter heated by laser-generated relativistic electrons in Ti wire target

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Warm dense matter (WDM) is of wide interest within the last decade both from fundamental physics and different applications points of views. In presented work we studied WDM generated up to 1 mm inside of a titanium wire target (of 50 μm diameter) irradiated by intense laser of PHELIX (Petawatt High Energy Laser for heavy Ion eXperiments) installation at GSI, Darmstadt. The proposition of thin metal wires to be used as a target allows to trace the isochoric heating of a matter deep into the target. Additionally, for such targets with reduced cross dimensions, a strong electrostatic sheath fields confine the electrons to the target which efficiently and homogeneously heat it to high energy densities. Among others diagnostics high spatial resolution X-ray K$\alpha-$spectroscopy of the selfemission (XES) was used to measure the temperature (with spatial resolution along the wire) of the matter isochorically heated by hot electrons propagating along the wire. The other diagnostic was a high-resolution point-projection X-ray absorption radiography (XAR) for diagnostic of the hydrodynamic expansion of the heated Ti wire heated. It was possible to distinguish surface target regions heated by mixed plasma mechanisms from those heated only by laser-generated hot electrons heating solid-density matter to temperatures up to 50 eV. There were two sets of experiments, results of the first one have been published [1], results of the second one are presented.

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Harmonic generation in inhomogeneous relativistic plasma

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We have presented the harmonic generation theory by the relativistic plasma resonance mechanism in an inhomogeneous laser plasma. A nonlinear current in the vicinity of the critical plasma density which is the source of the harmonic generation in the vacuum is calculated via the renormgroup symmetries method. Smooth power-law spectra of the the stationary radiation field are found and their characteristics are discussed. Dependences of the harmonic generation efficiency on the laser radiation incidence angle and on the plasma inhomogeneity scale are demonstrated. Transformation coefficients into higher harmonics for different laser-plasma parameters are calculated and comparison with non-relativistic theory is performed.

The study of ion acceleration by a femtosecond laser pulse of relativistic intensity in laser-plasma interaction with time-of-flight and Thomson parabola mass-spectrometers

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New analytical and numerical models of particle motion in the time-of-flight and the Thomson parabola mass-spectrometers, taking into account the edge effects of magnetic and electric fields, are presented and compared. We show that these edge effects introduce significant distortions in the particle trajectories. Using numerical simulations, the optimal parameters of the Thomson parabola mass spectrometer were determined, and the spectrometer with corresponding parameters was manufactured.

We present the experimental results on relativistic laser-plasma interactions of femtosecond laser pulse (Ti:Sapphire, wavelength – 800 nm, pulse duration – 45 fs, maximum intensity on target – $4{\cdot}10^{18}$ W/cm^2) with the front surface of a dense target. The experiments with low and high contrast levels of laser pulse are compared with the results of numerical simulations using the developed models of mass-spectrometers.

Single-cycle relativistic laser-plasma interaction: current status and prospective

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A possibility to shorten the laser pulse duration to the extreme level — a single cycle — oriented research in all possible domains, from laser science to a variety of applications, related to sub-cycle durations and energetic particles. Such pulses, dependent on carrier-envelope-phase (CEP), may on one hand, rectify the fields while interacting with the matter, and produce isolated sources of high energy photons or particles, on another hand, while focused tightly, they can produce unprecedented intensities which could be used further to test nonlinearities and even for boiling the vacuum. Single-cycle laser-plasma experiments come now into play in the relativistic level of 10^{19} W/cm^2, which provide the basis for future experiments at petawatt laser facilities. In this talk we will review the recent findings in experiments and modeling.

High-field effects in autoionizing states – high-order harmonics and the four-step model

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High-order harmonic generation (HHG) is an excellent tabletop source of coherent extreme ultraviolet and soft X-ray radiation. Since high-order harmonics are intrinsically generated in attosecond bursts, it is also opening a new domain of attosecond science. More recently, high-harmonic spectroscopy is developing as a unique tool for studying the electronic structure of atoms and molecules. The HHG process from most nonlinear media is well explained by the three-step model. However, for some media (such as the laser ablation plume), the intensity of a specific harmonic order could be enhanced by one to two orders of magnitude, compared with the neighboring harmonics that are generated due to conventional three-step process. This has been observed when the wavelength of a harmonic is in close proximity to an autoionizing resonance of the nonlinear medium.

In this talk, I will first review past works on HHG from laser ablation plume, and then present our recent studies on the electron quantum paths in the vicinity of an autoionizing state (AIS) of tin with mid-IR tunable driving fields. The present study shows that the resonant harmonic (RH) generation process involves the AIS for coherent harmonic emission at resonant energy via the microscopic response. Further, it has been revealed that the RH generation process involves the dressed-AIS for coherent harmonic emission at frequencies $\pm 2\Omega$ from the RH frequency (Ω represents laser frequency). As RH is an excellent candidate as a source for intense harmonics, the involvement of dressed states in HHG opens the perspective to expand the bandwidth of RH, thus opening the possibility to generate intense and ultrashort attosecond pulses, which would be useful for various applications in attosecond science.

X-ray radiation properties of plasma under interaction of femtosecond laser pulses with $\sim 10^{22}$ W/cm^2 intensities

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Study of radiation properties of solid dense plasma irradiated by ultraintense lasers has a great interest both from fundamental physics and different application point of views. Recently upgraded petawatt J-KAREN-P laser together with precise focusing technique delivers 35 fs laser pulses of 10^{22} W/cm^2 intensity into a micron-size focal spot on target. For such unprecedented intensities the application of high-resolution X-ray spectroscopy allows to investigate the ionization mechanisms and to measure the parameters of relativistic plasma from front and rear sides of moderate (Al) and high Z (Ti, Fe,) thin foil targets. Kinetic modeling of the spectra is used to estimate electron plasma density and temperature, demonstrating T_e ~2 keV for N_e ~5e22 cm^{-3} in the hottest emission region. Thus, it is experimentally demonstrated for the first time that the laser pulse of over 3e21 W/cm^2 intensity is absorbed neither in the solid density plasma nor in a pre-plasma of a common critical density, but in the matter of so called relativistic critical density. It is revealed how even small displacement of the target out of the optimal laser focus, as well the decrease in temporal contrast of the laser pulse, strongly reduce both the intensity of X-ray radiation and degree of plasma ionization. 2D PIC code simulations of femtosecond laser interaction with various materials are provided and compared with experimental results.

The study of electron acceleration in relativistic laser plasma through the analysis of trajectories

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Various research groups are experimenting with the production of electron beams as a result of laser pulse hitting the target. Based on these experiments, numerical simulations are performed using a PIC code. It turns out that the intersection of the electron trajectories causes their concentration to have sharp peaks. If they fall into suitable conditions in the field during the movement of these peaks, then they manage to escape from the plasma, with high energy. To describe these processes, a two-stage model was proposed: some laser accelerated electrons are able to leave the plasma in a narrow phase window of the moving wave interference pattern; then these electrons are directed along the normal to the target using the ponderomotive forces of the field. The mechanisms of electron acceleration to average values to the end are not clear.

The processing of numerical simulation had shown that the acceleration of electrons occurs as follows: a high intensity pulse incident on the target with preplasma excites plasma waves. Those waves' amplitude increases. Then it reaches the maximum value and the wavebreaking takes place. After the wavebreaking, the electrons are accelerated by the electric field of the plasma waves to energies of the order of 0.5 MeV (in the case of the intensity 10^{18} W/(cm^2)). Some of these electrons, caught in a successful phase of the electromagnetic field of the beatings of the incident and reflected laser waves, are captured by it and accelerated to energies of several MeV. It is established that most of the high-energy electrons are directed along the laser radiation beam reflected from the target. The transition to the angle of incidence of 60° shows an increase in the energy of the emerging electrons, which is caused by parametric processes. It was also noted that the growth of these energies occurs also with increasing length of the plasma gradient. That qualitatively coincides with the data obtained in the experiments.

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Ultra-high energy density plasmas using nanostructured plasmas

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As the laser technology continues its spectacular development, ever higher field intensities and power levels become accessible in laboratories. This opens new horizons for laser applications in ultra-bright sources of short wavelength radiation. At the same time, the laser pulse quality – like the contrast ratio – is greatly improved so that fine structured targets maintain their structure till the main pulse arrival. This opens new and unexpected possibilities for laser-plasma engineering towards new physics [1].

Nuclear fusion is regularly created in spherical plasma compressions driven by multi-kilojoule pulses from the world's largest lasers. Here we demonstrate a dense fusion environment created by irradiating arrays of deuterated nanostructures with joule-level pulses from a compact ultrafast laser. The irradiation of ordered deuterated polyethylene nanowires arrays with femtosecond pulses of relativistic intensity creates ultra-high energy density plasmas in which deuterons (D) are accelerated up to MeV energies, efficiently driving D–D fusion reactions and ultrafast neutron bursts. We measure up to 2×10^6 fusion neutrons per joule, an increase of about 500 times with respect to flat solid targets, a record yield for joule-level lasers. Moreover, in accordance with simulation predictions, we observe a rapid increase in neutron yield with laser pulse energy. The results will impact nuclear science and high energy density research and can lead to bright ultrafast quasi-monoenergetic neutron point sources for imaging and materials studies [2].

The efficient conversion of optical laser light into bright ultrafast x-ray pulses in laser created plasmas is of high interest for dense plasma physics studies, material science, and other fields. However, the rapid hydrodynamic expansion that cools hot plasmas has limited the x-ray conversion efficiency (CE) to 1% or less. Here we demonstrate more than one order of magnitude increase in picosecond x-ray CE by tailoring near solid density plasmas to achieve a large radiative to hydrodynamic energy loss rate ratio, leading into a radiation loss dominated plasma regime. A record 20% CE into $h\nu > 1$ keV photons was measured in arrays of large aspect ratio Au nanowires heated to keV temperatures with ultrahigh contrast femtosecond laser pulses of relativistic intensity. The potential of these bright ultrafast x-ray point sources for table-top imaging is illustrated with single shot flash radiographs obtained using low laser pulse energy. These results will enable the deployment of brighter laser driven x-ray sources at both compact and large laser facilities [3].

Ultrahigh-energy density (UHED) matter, characterized by energy densities $> 1\times10^8$ J/cm^3 and pressures greater than a gigabar, is encountered in the center of stars and inertial confinement fusion capsules driven by the world's largest lasers. Similar conditions can be obtained with compact, ultrahigh contrast, femtosecond lasers focused to relativistic intensities onto targets composed of aligned nanowire arrays. We report the measurement of the key physical process in determining the energy density deposited in high-aspect-ratio nanowire array plasmas: the energy penetration. By monitoring the x-ray emission from buried Co tracer segments in Ni nanowire arrays irradiated at an intensity of 4×10^{19} W/cm^2, we demonstrate energy penetration depths of several micrometers, leading to UHED plasmas of that size. Relativistic three-dimensional particle-in-cell simulations, validated by these measurements, predict that irradiation of nanostructures

at intensities of $> 1{\times}10^{22}$ W/cm^2 will lead to a virtually unexplored extreme UHED plasma regime characterized by energy densities in excess of $8{\times}10^{10}$ J/cm^3, equivalent to a pressure of 0.35 Tbar.

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Calculation of the absolute intensity of the x-ray source formed during irradiation of thin flat and structured Si foils by petawatt laser pulses

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It is well-known that position, shape, and intensity of spectral lines radiated by a plasma depends on its parameters such as electron density and temperature. It means that the plasma characteristics can be obtained by comparing experimental spectrum with calculated ones and nowadays the X-ray spectroscopy is one of the most powerful methods of plasma diagnostics. For most cases it is enough to use relative values of one of the mentioned above spectral line attributes to determine one or another plasma parameter. Due to the imperfection of spectrometric instruments distortions of the spectrum can be occurred, which may result in errors during the process of plasma parameters determination. That's why elimination of spectrum shape deformations associated with inaccuracy arising in measuring equipment provided by considering a hardware function of a spectrometric route is an actual problem in the field of the plasma X-ray spectroscopy.

We took in account influence of all parts of a spectrometric route used to record spectra of the plasma produced during irradiation thin flat and structured Si-foils by petawatt laser pulses generated by Vulcan PW laser facility. It allowed not only to correct the spectra shape, but also to obtain the real luminosity (photons/angstrom) of the X-ray source produced by the interaction of high-power laser pulses with solid targets. This value is an important parameter, which can give an opportunity to understand a distribution of the driver laser energy between different excitation processes take place inside a target.

Narrowband Compton scattering sources at high laser intensities

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Inverse Compton Scattering (ICS) is a valuable source of X- and gamma-rays for various applications in medicine, materials science and nuclear physics [1]. Growing interest in such sources is driven by the fast technological progress in both lasers and compact plasma wakefield based electron accelerators. Main advantage of ICS sources over, for example, commonly used bremsstrahlung sources is their ability to provide a very narrow bandwidth (<1%) photon spectrum, albeit with a very low conversion efficiency. One way to dramatically increase the total photon yield, is to increase the laser photon density, i.e. increase the laser pulse peak intensity. Unfortunately, scattering laser intensity is strongly limited by the fact that the generated spectrum can be nonlinearly broadened due to the ponderomotive force, and a bandlike structure can appear in the fundamental frequency as well as its harmonics [2,3] even for rather low values of laser pulse intensity. For example, such moderate laser pulse intensities as 10^{16} W/cm^2 already lead to broadening on the order of 2–4% considering 1 micrometer laser wavelength.

In this contribution, recent progress in nonlinear ICS will be discussed, analytical and numerical results of photon energy-angular spectrum calculation will be presented. Main focus will be on various methods to significantly enhance the photon yield of the photon sources mainly based on laser pulse frequency manipulation. By using novel geometrical method of spectrum calculation, we show the appearance of high-order caustics related to the catastrophe theory [4] in the case of laser pulses with linear chirp. It is also demonstrated, that the nonlinear broadening can be completely removed by proper nonlinear chirping [5,6]. We will compare different approaches for the photon spectrum calculation based on purely classical description with or without the radiation reaction and also based on strong-field quantum electrodynamics. Some of the fundamental aspects of interaction of single electron with electromagnetic waves will be addressed.

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Laser-plasma manipulation for achieving of femtosecond laser beam transportation of high performance

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Dissipative capillary discharges demonstrate possibility to form plasma wave guides for sufficiently long transportation of high intensity femtosecond laser pulses. For this reason, they are used in laser-plasma-accelerators of multi-Gev electrons. An additional laser heating of capillary plasma was proposed earlier to improve wave guiding and to diminish thermal load on the capillary walls. We combine firstly in this work 3D MHD code (MARPLE) and laser propagation code (INF&RNO) to get a tool for 3D simulations of complicated processes of mutual influence of plasma motion caused by the additional laser heating and laser beam propagation, influenced by redistribution of plasma density. We will show how such simulations help to choose optimal regimes for additional laser heating.

Plasma diagnostics based on the scattered harmonics radiation measurements

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There is a method of obtaining high-energy electrons based on the interaction of intense laser radiation with the target material in the plasma state. Such sources of high-energy electrons can be used to solve various problems of nuclear physics.

Today researches are carried out to find the most effective regime of high-energy electrons generation. The necessity of obtaining the information about the state of plasma interacting with laser radiation during this very interaction is obvious, which means firstly that we want to know plasmas' density distribution at any moment of time.

The following approach to this issue was developed. In the process of interaction of laser radiation with plasma, plasma waves arise. Which in term leads to the generation of optical harmonics during the interaction of plasma waves, scattered laser radiation, and initial laser radiation. Thus, from the picture of scattered radiation, one can estimate the state of plasma.

With the help of software developed during the research, we got the opportunity to analyze the spatial distribution of scattered radiation and its variation in time. After required analyzing done, we can obtain the necessary information on the state of the plasma, using the theoretical model describing the generation of the observed harmonics.

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Harvesting ultra-fast phenomena with the 10 PW laser system at ELI-NP

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By 2019 ELI-NP will host a laser system capable of producing 10 PW resulting in peak intensities of $I \sim 10^{23}$ W/cm^2. At such high intensities QED effects will play an important role in laser interaction with matter. As such it is hoped that new regimes of ultrafast phenomena can be discovered and exploited for the first time for fundamental and applied research at the interface of QED and nuclear physics. We will present an overview of the inaugural experiments planned for the nuclear physics station (E1) at ELI-NP. Foreseen Day-1 experiments will centre around the conversion of laser energy to gamma radiation for which efficiencies of up to 40% are predicted for powers >3 PW. Moreover, the intense and ultrafast laser-matter interaction within a short duration of 10s of fs could provide proton acceleration of >200 MeV if ultra-thin targets with d <150 nm are used, according to theory which predicts the emergence of the Radiation Pressure Acceleration (RPA) regime for such extreme conditions. We will present an overview of the planning stages with a focus on the implementation of the dedicated newly developed nuclear instrumentation.

Experimental studies on plasma physics and particle acceleration on PEARL facility

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We present an overview of recent experimental activities on the PEARL laser system (IAP RAS, Nizhny Novgorod, Russia). Scaled "laboratory astrophysics" experiments aimed to study the coupling between a high-amplitude large-scale magnetic field and high-velocity laser-plasma flows, could represent different astrophysical objects: solar arcs, astrophysical jets, accretion discs, which are characterized by complicated dynamics of plasma interaction with an ambient magnetic field. Experimental results of detailed studies of such plasma structures including development of different instabilities at the plasma-magnetic field boundary are presented.

Another set of experiments is devoted to study the laser-produced warm dense matter (WDM) with the use of high-power, high-contrast femtosecond PEARL laser beam. Heating efficiently solid density, or even compressed, matter has been a long-sought goal in order to allow investigation of the properties of such state of matter of interest for various domains. High-power lasers, pinches, and more recently Free-Electron-Lasers (FELs) have been used in this respect. Here we show that by using the femtosecond PEARL laser delivering 7.5 J in a 60 fs laser pulse an effective heating of a slab of solid density Al of 0.8 μm thickness at a temperature of 300 eV with minimal density gradients could be obtained. The characterization of the target heating is achieved combining X-ray spectrometry and measurement of the protons accelerated from the Al slab. The measured heating conditions are consistent with a three-temperatures model that simulates resistive and collisional heating of the bulk induced by the hot electrons. Such effective laser energy deposition is achieved owing to the intrinsic high contrast of the PEARL laser which results from the Optical Parametric Chirped Pulse Amplification technology it is based on, allowing to attain high target temperatures in a very compact manner, e.g. in comparison with large-scale FEL facilities.

Effect of the pre-plasma density profile on the molybdenum K-alpha X-ray emission generated by an intense femtosecond laser pulse

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We present evolutions of the energetic conversion efficiency of the K–α line X-ray emission at 17.4 keV generated by a laser beam tightly focused on a molybdenum bulk target. Experimental studies are performed employing 30 fs Ti:Sapphire laser pulses with peak intensities and contrast (with respect to an amplified spontaneous emission, ASE), which are varied within the ranges of 10^{16}–10^{19} W/cm^2 and 10^6-10^{10}, respectively [1]. The obtained results are compared with results of numerical simulations of the pre-formed plasmas created on the target surface by the ASE, which precedes the main fs pulse and starts 5 ns earlier.

The observed evolutions of the K–α emission efficiency (fig.1) show that the steepening of the electron density profile at laser pulse contrast lower than 10^9 begins to manifest itself at threshold laser intensities of $\sim 4\times10^{17}$ W/cm^2 almost independently on the contrast value. It points to the similar scale length of the electron density profiles of pre-formed plasmas in the experiments with fs pulses having the ASE contrast ratio lower than 10^{-9}. Such behavior can be explained by the fact that location of the main interaction region, which is situated near the laser reflection point, spatially matches the part of pre-plasma density profile, which is characterized by a smooth extended plateau (fig.2).

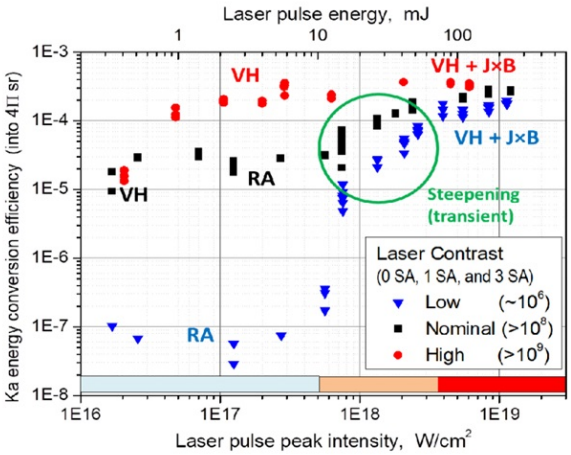

Figure 1: Energetic conversion efficiency of the Kα line emission from Mo target in dependence on the laser pulse peak intensity observed at different pulse contrast with respect to ASE. The pulse contrast is varied by changing the number of saturable absorbers (SA) incorporated into the laser amplification chain. RA, VH, and J×B indicate the dominant laser-plasma interaction mechanism: resonant absorption, vacuum heating, and ponderomotive heating, respectively.

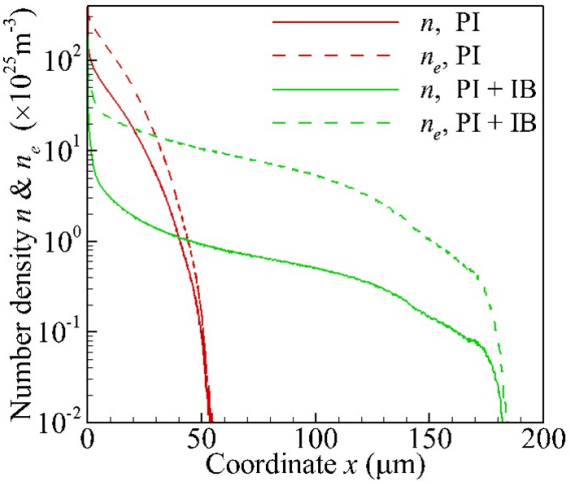

Figure 2: Density profiles of electrons, n_e, and heavy Mo particles (atoms and ions), n, in the pre-plasma plume, which are simulated for the ASE pedestal of 10^{10} W/cm^2 intensity and 5 ns duration, with and without the inverse Bremsstrahlung (IB) absorption. PI denotes photoionization.

Our original 1-D simulation code is based on the Direct Simulation Monte Carlo method (DSMC) extended for plasma flows using the assumption of local neutrality of plasmas. The results show that the characteristic plateau is formed on the pre-plasma density profile under ASE intensities exceeding 10^9 W/cm^2 due to an intense inverse Bremsstrahlung absorption of ASE radiation within the external layer of the plume of ablated particles. This layer can screen more than 90% of the incident ASE intensity. The plasma expansion partly inhibits the rise of plasma temperature caused by the radiation absorption and favors homogenization of the physical conditions within the internal layers. Nevertheless, the plasma density exhibits a steep rise in the vicinity of target surface (at $\sim$1 μm distance) because of the intense inflow of particles from the Knudsen layer. This region is characterized by a steep fall of the electron density from the solid to approximately the critical electron density (n_{ec}=1.7$\times 10^{21}$ cm^{-3}) with the scale length much smaller than the laser wavelength, L$\ll \lambda$. At the distance of several μm from the solid target, where the electron density is of $\sim 10^{20}$ cm^{-3}, the steep fall transforms into an extended smooth plateau (shelf) with L$\gg \lambda$, which ends rather sharply at distances exceeding tens and hundreds microns. Considering the subsequent ionization of the pre-plasma particles in the strong electromagnetic field of the rising edge of ultra-intense fs pulse, such shape of the pre-plasma density profile allows us to explain the observed independence of the threshold intensity of steepening of the electron density profile at the pulse contrast values below 10^9.

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Perspectives for high-power laser-driven nuclear physics

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High-power, short pulse lasers during the last decade have proven their capacity for the production of high energy γ-rays, charged particles and neutrons, and for inducing a variety of nuclear reactions. With laser and optical technology continuously improving and next-generation (multi-) PW laser facilities like ELI (Extreme Light Infrastructure) on the horizon, nuclear reaction studies based on high-power lasers will open new perspectives for various applications in nuclear physics research, in particular for nuclear astrophysics. Laser-driven ion acceleration with adequately tailored subsequent interactions will complement the portfolio of nuclear reactions so far available from conventionally accelerated particle beams. This presentation will give an overview on selected topics that will come into experimental reach with soon available high-power laser systems, pushing into new regimes of laser-driven particle acceleration.

The unprecedented density of laser-accelerated ion bunches, reaching solid-state density and thus many orders of magnitude denser than any ion bunch accelerated by conventional acceleration schemes, will allow for a novel nuclear reaction mechanism ("fission–fusion"), capable of generating extremely neutron-rich isotopes in the vicinity of the astrophysically important "waiting point" of the r-process heavy-element nucleosynthesis path at the waiting point of the magic nucleon number N = 126.

A second field of research, which will greatly benefit from synergies between nuclear physics and high-power laser-matter interaction, is the study of nuclear reactions and nuclear excitation/de-excitation processes under hot plasma conditions, as they are found in the inner cores of planets or stars. Thus experiments at high-power lasers can turn into a nuclear-astrophysics laboratory with PW-class laser systems. Plasma-induced modifications of nuclear lifetimes or reaction rates will have considerable influence on our understanding of nuclear processes under stellar conditions. The talk will present selected examples for such effects.

In conclusion, high-power, short-pulse lasers bear the potential to open up new vistas in particular for nuclear astrophysics, complementary to large-scale conventional particle accelerator centers.

Electrons ejection and acceleration in plasma waves in the relativistic laser-plasma of solid targets

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The electron bunches with high energy and charge may be generated in the plasma via different mechanisms of laser acceleration. In this paper we present the experimental research and numerical simulations of the mechanisms of laser energy absorption in nearcritical plasma at an intensity around 5×10^{18} W/cm^2.

In our experiments we used Ti:Sapphire laser system (wavelength – 800 nm, repetition rate – 10 Hz, maximal pulse energy – 30 mJ, minimal pulse duration – 45±5 fs and maximal intensity on target – $5\cdot10^{18}$ W/cm^2, contrast on ps time scale - 10^{-8}). The laser radiation was focused obliquely by the off-axis parabolic mirror (F∼5 cm) onto the metal or plastic target. To create pre-plasma layer on the target surface we use Nd:YAG laser (wavelength –1064 nm, repetition rate – 10 Hz, maximal pulse energy – 200 mJ, pulse duration – about 6 ns and maximal intensity on target – 10^{12} W/cm^2). Varying delay between the main pulse and the pre-pulse in the range from -50 ns to 10 ns and type of the target we can change pre-plasma properties significantly. Bremsstrahlung from hot plasma was detected by scintillation detector based on NaI crystal (1 to 7 cm thick). The electron beam was detected with the Lanex scintillator and a CCD matrix. Neutrons produced in the photonuclear reaction in the secondary D_2O and Be targets were detected by ten ^{3}He-counters CHM-18 with a polyethylene moderator, the signal from which, after amplification, was recorded by a digital oscilloscope in each laser shot.

We made interaction simulations using the fully relativistic 3D3V PIC code "Mandor", reduced to the 2D3V within most of the our calculations. The simulation box size was 46 μm (x) × 34 μm (y), spatial resolution was $\lambda/100$ and total number of particles was 10^8. Temporal resolution was 3×10^{-3} fs. The planar foil target consisting of cold ions and electrons. A p-polarized laser pulse with duration of 50 fs (FWHM) and central wavelength of 1 μm entered the simulation box.

In the experiment, the generation of the electron bunch with energies up to 6 MeV was obtained. In this case, the beam divergence is 0.1 radians. The beam has a large number of electrons with an energy of more than 1.7 MeV. This is confirmed by the neutron yield from the photonuclear reaction of gamma radiation obtained from a beam in a tungsten converter with a beryllium target.

We have clarified mechanisms of laser energy absorption and electron bunch generation in plasma via comparison of numerical simulation and experimental analyses of optical radiation and electron beams from plasma. It is shown that at the first stage the electrons accelerate to energies of the order of 1 MeV due to the breaking of plasma waves. The main mechanisms of plasma waves excitation are parametric instabilities (TPD and SRS). In the second stage, the electrons are injected into the field of the reflected laser pulse and are accelerated. With a relatively short plasma this corresponds to the VLA, and we have strongly divergent electron beam. The optimal acceleration regime is realized if the plasma has a short gradient near the critical density and an extended low density corona.

The reflected laser pulse forms a channel into which injected electrons are accelerated, forming a collimated beam. This corresponds to the DLA.

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Laser pulse diagnostics via direct particle acceleration

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Nowadays laser facilities allow achieving PW peak powers of laser pulses. Extreme intensities of such laser beams focused on the diffraction limit require new methods of diagnostics of laser parameters. In our report, based on the direct acceleration of particles (such as protons and electrons) we propose new technique as one of the possible methods for these purposes. This approach demands laser-particle interactions to be dominant as compared to the interaction between particles through the plasma fields; such condition can be realized in the case of ultrathin nanofoils or rarefied gases. We use Stratton-Chu integrals to model laser beam focused on the focal spot with a size near to the diffraction limit, when extraordinary intensities could be attained, by off-axis parabolic mirror used in real experiments. Neglect of the effects of particle interactions simplifies the basic equations to equations of test particle motion in relativistic form with a Lorence force. We have shown a good accuracy of ponderomotive approximation for protons in the entire range of considered intensities (up to 10^{23} W/cm^2). By using our approach, we have carried out a number of calculations varying laser parameters, such as the peak intensity, focal spot size and spatial-temporal profiles and shown applicability of proposed method for a number of laser pulse diagnostics.

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Section 2: Ultrafast phenomena in condensed matter and ionized gases

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Program committee:
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Sergey Popruzhenko (Max Planck Institute for the Physics of Complex Systems, Germany)
Stanislav Bezhanov (LPI, Russia)

Scope
Optical properties of heated solid conductors
Instabilities and high-frequency phenomena in photoionized plasma
Non-linear phenomena in nonequlibrium plasma and metals
Kinetics of rapidly heated electrons in metals and plasma

The behavior metals under ultrafast loading driven by femtosecond laser

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The results of experimental investigation of ablation and shock-wave phenomena in metals under ultrashort loads driven by femtosecond laser pulses are presented. Ultrafast interferometry was used for diagnostics of movement in a picosecond range of frontal and rear surfaces of thin film samples. The evolution of ablation plume and elastic-plastic shock waves has been studied. The data on the HEL and spall strength values have been obtained in strongly metastable states close to ultimate values of shear and tensile stresses.

Signatures of attosecond-scale electron dynamics in terahertz and higher order Brunel harmonics

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Terahertz (THz) frequencies lie at the long-wavelength limit of optical range (wavelengths around 100 μm) and are extremely useful for many applications, mostly biological ones, since transitions between rotational and vibrational levels of complex molecules lie in this range [1]. On the other hand, because of long wavelength and thus long oscillation period (around 1 picosecond) it is quite unusual to expect that THz radiation gives information about the dynamics happening on much lower times scales, for instance in femto- and atto-second ranges [2]. The latter is typically studied with help of so called high-harmonic generation (HHG), a process which is based on ionization of electrons from atoms and their subsequent dynamics, including their return to the core [2]. This return provides very high-order coherent harmonics in the range of tens and hundreds eV — the range which is inaccessible by the other means.

However, even without return to the core, electron ionization and subsequent dynamics in atoms produce harmonics of relatively low order (up to few tens of eV) [3]. Although fully free electrons (pre-ionized plasma) do not create any harmonics, the process of ionization followed by the subsequent dynamics in the field does, since ionization changes the refractive index in a field-dependent manner and thus creates nonlinearity. The corresponding radiation is referred to as Brunel harmonics. It was also realized, that in the fields which break the space-reflection invariance of the ionization-created macroscopic plasma currents, 0th Brunel harmonic can appear which typically cover the THz and sometimes mid-infrared (MIR) frequency range [1]. The typical position of low-frequency cut-off is determined by the free-electron current decay and is typically around 0.1 THz, whereas highest generated frequencies are determined by the pulse duration and the broadening due to propagation effects and can lie in MIR range.

Here, we demonstrate several ways allowing to extract ionization dynamics, often on the attosecond time scale, from the 0th Brunel harmonic. First, we note that if we decrease the intensity starting from the one around 100 TW/cm^2 (in the tunnel ionization regime), at certain point in intensity the inverse ionization rate approaches the duration of a single cycle of the pump pulse. At this point the transition from tunnel to multiphoton regime takes place. The signature of this transition can be seen in the amount of THz Brunel radiation per single ionized electron. Namely, if we are in the multiphoton regime, nonlinearity becomes too slow and the energy per single ionized electron is drastically reduced [4]. The same mechanism leads also to depletion of higher-order Brunel harmonics.

As a second example, we consider THz radiation from single color fields where the symmetry is broken by the intra-atomic excitation of Rydberg states in course of ionization process (so called Freeman resonances) [5]. Freeman resonances take place also if the intensity is scanned as sudden peaks of ionization rate at certain intensity. In a long enough pulses we may consider the growth and fall of the intensity at the pulse edges as the intensity scan. In this case, narrow Freeman resonances are translated to ionization peaks which are narrow in time. It may happen that the duration of such peaks can be even shorter than the period of a single oscillation of the driving pulse. This creates a symmetry breaking mechanism which also leads to low-frequency generation [6].

Finally, so called attoclock shows up in the THz radiation of certain waveshape [7]. Attocklock was introduced as a electron-detection technique allowing to measure tunneling time of electron as it is ionized by the field. To do this, electron is ionized in a field with polarization close to circular. The driver electric field servers as a hand of a clock, and this clock starts at the moment when the electron is ionized. The typical times which are inferred from the attoclock are around 100 attoseconds. We found a 2-color driver waveshape, which consists of two co-rotating circularly polarized electric fields, which leads to emission of 0th Brunel harmonics. Remarkably the resulting Brunel radiation is linearly polarized, and the polarization direction is rotated by $\tau\omega_0$ where τ is the ionization delay and ω_0 is the fundamental driver frequency [7].

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Nonlinear transmission and reflection of a strong terahertz pulse by a metal film

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Nonlinear interaction of the powerful terahertz radiation pulse with metal nanofilm is investigated. For currently achievable experimental values of THz field strength it is possible to observe significant changes in reflectivity and transmissivity of metal films, which is demonstrated by the analytical model and numerical simulation using 30 and 100 nm aluminium films. Nonlinear heating of the conduction electrons during absorption of the 500 fs, $30-90$ MV/cm THz pulse creates the excited nonequilibrium state. For these conditions, pulse duration may be of the same order of magnitude as the sample electrodynamic and thermal temporal scales. The effective electron collision frequency in this nonequilibrium state increases multiple times compared to the one for the cold film. This causes an order of magnitude increase of film transmissivity, as well as noticeable change in reflectivity.

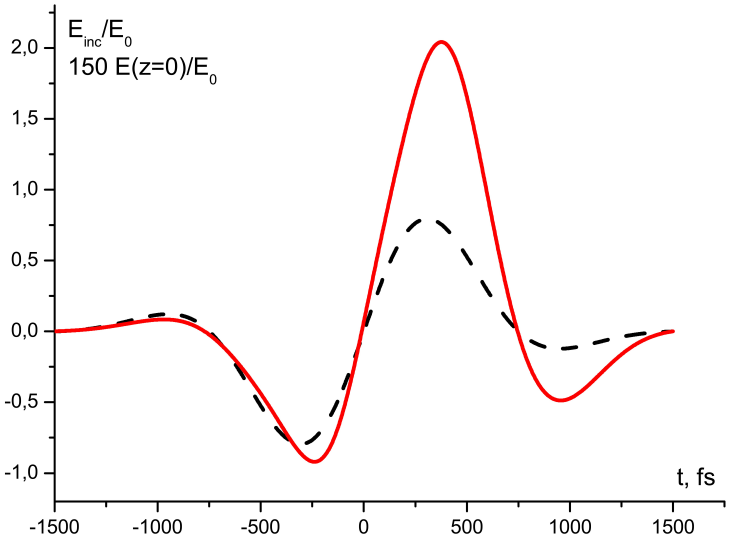

Figure 1: Temporal dependence of the field in the film (red line), compared with the incident pulse shape (black dashed line) for $d = 30$ nm film irradiated by $E_0 = 30$ MV/cm pulse.

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Plume splitting and oscillatory behavior in transient plasmas generated by high-fluence laser ablation in vacuum

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We present an overview of studies performed in our group over the last decade on the characterization of transient plasma plumes generated by laser ablation in various temporal regimes, from nanosecond to femtosecond [1-7]. Optical (fast gate intensified CCD camera imaging and space- and time-resolved emission spectroscopy) and electrical (mainly Langmuir probe) methods have been applied to experimentally investigate the dynamics of the plasma plume and its constituents. Peculiar effects as plume splitting and sharpening or oscillations onset have been evidenced at high laser fluence. New theoretical approaches (based on non-differentiability and Scale Relativity Theory) have been developed to qualitatively and quantitatively account for the experimental observations.

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Dipole structure of terahertz radiation in the interaction of a laser pulse with clusters

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The theory developed earlier in [1, 2] for quadrupole radiation of THz waves under laser-cluster interaction is generalized to the case of the dipole mechanism of THz field generation. It is shown that the dipole structure of THz radiation is realized for small-size clusters with a sufficiently high electron collision frequency. The results of the theory of penetration of laser radiation into a cluster with dimensions much smaller than the skin depth are presented and the spatial distribution of the laser field inside the cluster for an arbitrary density of free electrons is found. It is shown that the electric field of laser radiation is appreciably amplified in a supercritical cluster plasma if the laser frequency is close to the frequency of the dipole or quadrupole mode of a plasma sphere. The excitation of low-frequency electromagnetic fields in a cluster under the ponderomotive action of laser radiation on free electrons is considered and the spatial distribution of low-frequency electromagnetic fields in the cluster and vacuum is calculated. The results obtained for vacuum low-frequency fields are analyzed at large distances from the cluster in the wave zone. The angular, spectral, and energetic characteristics of THz radiation are studied. It is established that the spectrum of THz radiation depends substantially on the density of free electrons. For a low-density cluster plasma, the emission spectrum has sharp peaks at the frequencies of dipole and quadrupole oscillations of the plasma sphere. These peaks are associated with the so-called leaky modes that are excited in the cluster under the action of laser radiation and can be emitted into the ambient medium. As the density of free electrons increases, these peaks gradually disappear and, in the case of an supercritical electron density, are replaced with a wide peak at a frequency comparable with the reciprocal of the laser pulse duration. It is shown that, for small clusters with sufficiently frequent electron collisions, the angular energy distribution has a dipole structure and THz waves are emitted mainly at an angle of $\pi/2$ with respect to the propagation direction of the laser pulse. The total energy of THz radiation is calculated, and its dependence on the density of free electrons is analyzed. It is shown that the energy of THz radiation is maximal for a dense cluster plasma under the resonance conditions in which the frequency of laser radiation coincides with the eigenfrequency of a dipole or quadrupole mode of the plasma sphere. It is established that the dipole mechanism of THz emission prevails in small-size clusters with frequent electron collisions. The spatiotemporal structure of the electromagnetic field in a THz pulse is studied. It is shown that the profile of the THz signal depends substantially on the density of free electrons. For a low-density cluster plasma, the field of the THz pulse oscillates at the frequencies of the dipole and quadrupole modes of the plasma sphere, and its duration is determined by the damping rate of the corresponding mode and can reach a few picoseconds. For a supercritical electron density, the THz signal contains only two oscillation cycles and its duration is comparable with the duration of the laser pulse. Estimates show that, in the interaction of femtosecond laser pulses with clustered gases, high-power THz pulses can be generated with a rather high conversion efficiency.

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Nonthermal phase transitions in silicon and antimony: scaling up ab-initio atomistic simulations

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Intense femtosecond-laser pulses are able to induce ultrafast structural phase transitions in different materials along pathways that are inaccessible under thermodynamic conditions. In order to investigate the nonthermal motion of atoms in Si and Sb upon femtosecond laser excitation we performed ab-initio molecular-dynamics simulations on laser-excited potential energy surfaces using our code CHIVES (Code for Highly excited Valence Electron Systems). In the case of Si were able to characterize particular signatures of nonthermal phenomena like thermal phonon squeezing [1], fractional diffusion [2] and anisotropic energy flow [3]. For Sb, we obtained the decay paths of the coherent A1g-phonon as coming mainly from four-phonon scattering processes [4] and we studied the possibility of reversing the Peierls distortion.

In order to extend the ab-initio description to larger length scales to study, for instance, nucleation phenomena, we succeeded in developing analytical interatomic potentials for laser-excited silicon and antimony, which depend on the electronic temperature. The potentials are based on polynomials and the coefficients are fitted to simulations performed with CHIVES under different conditions. For each material, we generated a data base of many hundred thousand possible potentials and selected the optimal ones by minimizing simultaneously the error with respect to the ab-initio runs and the number of coefficients. Effects like bond softening in the presence of hot electrons are taken into account and are reproduced by the analytical potentials. With the help of these potentials we were able to perform large-scale simulations and study nucleation dynamics during nonthermal melting of Si and the laser induced "undoing" of the Peierls distortion in Sb. We found anomalous heterogeneous nonthermal melting in Si, driven by bond softening. Moreover, in the case of Sb films we obtained that the inverse Peierls transition is mostly a cooperative phenomenon and that incoherent electron-phonon scattering helps to stabilize the simple cubic phase. The possibility of using laser-excited analytical interatomic potentials to describe laser processing will be discussed.

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Generation and amplification of THz radiation in plasma channel formed in gas by high-intensity laser field

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New approach to construct the source of THz radiation is discussed. It is based on the formation of induced THz currents by two-color femtosecond optical pulse which are supposed to be injected and amplified in the extended nonequilibrium plasma channel by the mechanism proposed in [1]. The additional gain of the THz signal in plasma can ultimately lead to construction of more efficient THz sources. The optimal parameters of the pump two-color pulse are investigated and possible realizations of such an amplification scheme are discussed.

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Cycle-averaged effects in ultrafast high-intensity laser interaction with electrons of wide-band-gap solids: the approximation of low collision rate

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Solids transparent to low-intensity laser radiation deposit laser-pulse energy only via nonlinear excitation of their electron sub-system, e.g., by multiphoton absorption. However, the valence electrons of the solids intensively interact with other particles of those quantum systems, and that interaction leads to significant challenges in theoretical and experimental studies. For example, two types of laser-driven electron dynamics compete in the solids: collision-driven chaotic dynamics and coherent laser-driven oscillations. In a majority of simulations, an *a priori* assumption is made regarding the type of dominating dynamics to fit experimental data. The fitting delivers 1-fs electron-particle collision time. It is significantly smaller than duration of a single optical cycle at infrared laser wavelengths and is favorable to assume the collision-driven electron dynamics dominates in the ultrafast high-intensity laser-solid interactions. This approach leads to extensive use of high-collision-rate models (e.g., the Drude model of collision-dominated electron dynamics) coupled with zero-collision-rate models (e.g., the Keldysh formula for the photoionization rate). This internal contradiction has been recognized, but has not received a proper fix.

Moreover, recent publications of ultrafast strong-field solid-state effects suggest that the laser-driven electron oscillations dominate in wide-band-gap crystals. In this connection, it is highly reasonable to build a theoretical approach based on the assumption that the coherent laser-driven oscillations are the major type of the laser-driven electron dynamics. High intensity (10–100 TW/cm^2) is a specific feature of those ultrafast laser-solid interactions. Therefore, time-dependent electric field of the laser pulses substantially distorts the electron sub-system, and the regular perturbative approaches are not reliable.

We discuss an approach to build an analytical model of the ultrafast laser-driven inter-band and intra-band electron excitations that: (a) is non-perturbative, i.e., is valid at high intensity; (b) does not use a quasi-monochromatic approximations for laser radiation, i.e., can treat the broad-band ultrashort laser pulses; (c) does not *a priori* assume huge electron-particle collision rate; and (d) is simple enough to serve as a replacement of the contradictory rate-equation models. That model can be developed by averaging the oscillatory effects over single oscillation cycle. In this case, cycle-averaged energy of the oscillations (i.e., ponderomotive energy) can be correctly introduced and properly accounted for.

We begin with analysis of ultrafast modification of energy bands of a wide-band-gap crystal by the ponderomotive energy. It is shown that a crystal with initial direct-gap energy bands interacts with a high-intensity ultrashort laser pulse as an indirect-gap crystal. Formation of the transient indirect-gap energy bands leads to generation of specific cycle-averaged photocurrent in dielectrics and suppression of nonlinear absorption at the leading edge of the laser pulse. Second, we discuss the cycle-averaged photoionization rate for the inter-band transitions between the indirect-gap transient energy bands driven by envelope of an ultrashort laser pulse. It is shown that the dynamics of the rate substantially departs from the traditional predictions of the Keldysh formula. Formation of

an abrupt ionization front is predicted. Finally, we replace the traditional high-collision-rate Drude model of intra-band conduction-electron dynamics with the low-collision-rate Vinogradov model. Contributions of several valence bands are taken into account. The Drude and the Vinogradov models are compared by numerical solving a rate equation for conduction-electron density coupled to an equation for energy absorption by the conduction electrons. Substantial qualitative difference between those approaches is discussed.

The role of nanoparticles in optical breakdown of liquids: impact on the main parameters of plasma and sound, values of radiation-chemical yeild of stable and radical photolysis products

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The report will present the effect of gold nanoparticles on the main parameters of plasma and sound during optical breakdown, the radiation-chemical yields of stable and radical photolysis products in water and organic solvents. Gold nanoparticles were obtained by laser ablation, with an average diameter of 16 nm. Optical breakdown in colloidal solutions of such particles was carried out using an Nd:YAG laser (1064 nm, 10 ns pulse with energy up to 2 mJ, frequency 8 kHz). Laser radiation was focused inside the liquid through a window and moved along a linear trajectory of 20 mm in length at a speed of 3000 mm/s. The calculated diameter of the laser beam at the focus was 30 μm, which corresponds to a density of the laser in a liquid of 140 W/cm^2. A bright line from the plasma appeared in the experimental cell 2–3 mm above the surface of the window. Amperometric sensors for molecular oxygen and molecular hydrogen were integrated in the experimental cuvette, a highly sensitive film sensor was used to record the acoustic oscillations induced by laser radiation, and a luminescence of the plasma produced during optical breakdown occurred with a CCD camera. The images received from the CCD camera were processed with the help of the LaserImage program developed by us.

The effect of the concentration of gold nanoparticles (10^9–10^{11} pieces/ml) on the luminescence characteristics of the region of optical breakdown in the visible region of the spectrum was studied. In deionized water without nanoparticles, the probability of optical breakdown is close to 1%. The probability of optical breakdown per laser pulse increases exponentially with increasing nanoparticle concentration to 2×10^{10} pieces/ml and approaches 90%. A further increase in the concentration of nanoparticles leads to a monotonic decrease in the probability. The number of optical breakdowns of the laser pulse observed in one track behaves in a similar way. The integrated intensity of a single optical breakdown decreases monotonically with increasing nanoparticle concentration. The dependence of the average distance between breakdowns in the track on the concentration of nanoparticles was investigated. It is shown that the average distance between breakdowns in one track varies nonmonotonically with increasing nanoparticle concentration, details will be presented in the presentation. Also, the report will present data on the effect of energy in the laser pulse on the luminescence characteristics of the optical breakdown region. The effect of the concentration of nanoparticles and energy in a pulse on the intensity of acoustic oscillations induced by laser radiation will be presented in the presentation. The influence of the concentration of nanoparticles and energy in the pulse on the radiation-chemical yields of 1. hydroxyl radicals, 2. hydrogen peroxide, 3. molecular oxygen, 4. molecular hydrogen will be presented in the presentation too. Also, the effect of optical breakdown plasma on the formation of molecular products in organic solvents (ethanol, propanol–2, butanol–2 and diethyl ether) will be shown.

The work is supported by the Grants of the President of the Russian Federation (MD-3811.2018.11 and MK-3606.2017.2), RFBR grants 16-02-01054_a and 18-52-70012 e-Asia_a.

Role of hydrodynamic mechanisms in formation of laser-induced periodic surface structures

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Hydrodynamic instabilities usually develop on slow time scales and are not associated with ultrafast phenomena. However some properties of the laser-induced periodic surface structures (LIPSS) or ripples, appearing upon femtosecond laser irradiation of metals, can only be explained if the optical mechanisms of the LIPSS formation are extended by hydrodynamic processes. Here the combined model of the ripple formation is suggested: the initial small perturbation of the electron temperature on the laser-irradiated surface provides an initial perturbation, which will be amplified by hydrodynamic instabilities. The question how the electron temperature profile manifests itself in a periodically-modulated surface morphology in such a short time (less than 1 nanosecond) is addressed. Estimations made on the basis of different hydrodynamic instabilities allow us to sort out several mechanisms, which can bridge the gap between the temperature and the surface profile formation.

Numerical modeling of energy relaxation in molybdenum thin films on soda-lime glass upon irradiation by picosecond laser pulses

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Laser processing of thin metallic films is widely used in industry for photovoltaic applications and electronic component manufacturing [1]. However, achieving a high processing quality is still challenging and requires further investigations with addressing strong sensitivity of the laser ablation process to laser irradiation parameters. From fundamental point of view, laser irradiation of thin films enables to study subtle effects such as rapid electron excitation, the transient change of optical properties, a thickness-dependent electron-phonon collision rate [2], stress generation and film removal mechanisms.

In this work, a numerical code based on two-temperature model (TTM) was developed for the case of a thin molybdenum film deposited on soda-lime glass irradiated by short laser pulses of 10 ps. Ensuring the energy conservation of the numerical solver, melting threshold fluences were calculated for different thicknesses of Mo films. The simulation results show good agreement with experimental data [3]. Several temperature dependencies of the thermal conductivity and/or heat capacity of soda-lime glass, existing in the literature, were tested. It was found that damage threshold depends not only on pulse duration and film thickness but is also strongly affected by thermal conductivities of the film and underlying substrate.

The film spallation phenomenon will be discussed based on calculations of fractions of molten molybdenum as a function of depth for the cases of incomplete melting and estimations of laser-induced stresses.

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Ablation-cooled material removal with ultrafast bursts of pulses

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Ultrafast lasers allow thermal damage-free ablation irrespective of the material type. However, this is a slow and inefficient process, because enough time has to pass between subsequent pulses for the material to cool down to avoid heat buildup. We recently demonstrated ablation-cooled laser-material, whereby ultrafast pulses are sent so quickly one after another (within 100's ps of each other) that there is no time for heat to diffuse away from the processing region. In this regime, the ablation rate increases by orders of magnitude so that the ablation process becomes the dominant heat removal mechanism and the rest of the target material remains cool and without damage.

Laser action on bulk or thin targets: duration effects

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Laser action on targets with different geometries (1D-2D-3D, bulk targets, films, laminates, ablation in liquid) by pulses with different pulse durations (from 10s of fs to nanoseconds), with wavelengths from IR to soft and hard x-rays, by Gaussian and non-Gaussian beams is considered. Physical processes covering multiscale hierarchy of time durations up to 100s of nanoseconds are described. Formation of metasurfaces is analyzed. It is demonstrated how using splitting of the processes in time allows to create a hybrid approach combining electrodynamic, two-temperature 1D hydrodynamics, 2D hydrodynamics (bulk and thin shell approximation), molecular dynamics (MD), Monte-Carlo, and smoothed particles hydrodynamics (SPH) codes: output of the one code serves as the input for the next one. The approach includes interference and absorption of electromagnetic waves, electron-ion coupling, thermal transport, real equations of state for condensed matter, first order phase transitions, and EAM (embedded atom model) interatomic potentials. By this approach we overcome severe resource limitations of modern multiprocessors supercomputers (memory and flop/s limits) obstructing direct solution of laser problems at real spatiotemporal scales; e.g. to describe microbumping of a film 0.3 micron thick on a substrate more than 100 billions of atoms in MD simulation are necessary - while today limit at the best supercomputers is $\sim$ 1 billion.

Authors (NAI, YVP, VAK) acknowledge the financial support of FASO RF (project № 0033-2018-0004) for developing, checking, and running the numerical codes and authors (NAI, VVZ, YVP, VAK, VVS, KPM, DKI) acknowledge support from Russian Foundation for Basic Research (RFBR) (grant № 16-08-01181-a) for creating and developing of physical models.

Evolution of nonequilibrium state of the high-temperature superconductor after femtosecond laser pulse, studied with numerical simulation

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The non-equilibrium state of the high-temperature superconductor after action of ultrashort laser pulse is studied. The evolution of the non-equilibrium state is simulated with numerical solution of kinetic equations for quasiparticle occupation numbers. We show the simulation results for laser pulses of different duration and envelope shape, demonstrate the relaxation and cooling of excess particles, and discuss the role of the superconductor temperature. The characteristic relaxation times are discussed. The analytic transformations used in the simulation are shown. The work is supported by Russian Foundation for Basic Research, grant № 17-29-10024.

Attosecond pulse formation in active medium of a plasma-based x-ray laser, dressed by a strong optical field: analisis and optimization

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The plasma-based x-ray lasers are unique laboratory sources of electromagnetic radiation, allowing for generation of high-energy (up to several mJ) picosecond pulses in the wavelength range 3–50 nm [1]. However, the picosecond duration of the pulses limits their applications for study and control of the ultrafast optical processes, unfolding on femto– and attosecond time scales.

Recently, it was shown that a plasma-based x-ray laser (generating at the transition n=1↔n=2 of hydrogen-like ions) used as an amplifier allows to produce an attosecond pulse train from a picosecond (or sub-picosecond) pulse of an incident x-ray field [2]. For this purpose, an active medium of the x-ray laser should be irradiated by a strong optical laser field, providing sub-laser-cycle Stark shift of the upper lasing energy level [3], and thus leading to spectral broadening of the amplified x-ray field.

In the present work we continue to study this problem and obtain the analytical solution for the output x-ray field. Analyzing the obtained solution, we identify the two regimes of the pulse formation: (a) the "overmodulation" regime, which corresponds to the amplified x-ray field consisting of only three phase-locked spectral components with comparable amplitudes; and (b) the "spectral broadening" regime, corresponding to broader spectrum of the output x-ray field. We study these regimes both analytically and numerically (by solving the self-consistent set of Maxwell-Bloch equations taking into account spontaneous emission) and find the optimal values of (i) the modulating optical field (intensity and wavelength) and (ii) the active medium (depth and concentration of free electrons and active ions), for both regimes using different optimization criteria. The feasibility of sub-fs pulses formation at wavelength 13.5 nm is shown for the x-ray laser based on Li^{2+} hydrogen-like ions [4], dressed by a laser field with wavelength 0.8–2 μm and intensity 10^{14}–10^{15} W/cm^2.

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High harmonic generation by atoms and atomic ions near cut-off

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The yield of spontaneous photons at the tunneling ionization of atoms by intense low-frequency laser radiation near the classical cut-off is estimated analytically within the three-step model. Bell-shaped dependence in the universal photon spectrum is explained qualitatively. At the tunneling ionization of atoms by intense low-frequency laser radiation of linear polarization the first (quantum) step is the penetration of an electron through slowly oscillating potential barrier. The second (classical) step is the possible rescattering of this electron after some part of laser period on an atomic core. The possibility of this process is determined by the value of laser phase when an electron leaves the potential barrier. During the second step electron achieves the energy of the order of ponderomotive energy U_p from the laser field that is much larger than the atomic ionization potential I_p. The third (semi-classical) step is the emission of high-energy spontaneous photon when an electron returns into its initial ground atomic state. Maximum of photon energy is $E_{max} = N_{max}\omega = 3.173U_p$ [1]. Of course, alternatively, on the third step this electron can be elastically scattered by atomic core [2]. The energy distribution of electrons rescattered by their parent ions and emitted near the classical cut-off is considered analytically in Ref. [3]. The goal of this talk is to investigate qualitatively the spontaneous photon yield near N_{max}. High harmonic spectrum is of the bell-shaped form. Of course, this is envelope of the harmonic spectrum consisting from odd harmonic numbers N. Beside of this, the maximum of this dependence is shifted remarkably to the less value than classical cut off ($\hbar N_{max}\omega = 3.173U_p$). The reason is strong decrease of recombination cross section with the photon energy. These conclusions are in a good agreement with the experimental data [4]. Some details of this talk can be found in Ref. [5].

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Generation of sub-femtosecond electron bunches upon laser pulse propagation through a sharp plasma boundary

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Investigation of the generation of electron bunches when a relativistic-intensity laser pulse propagates through a sharp plasma boundary revealed the main features of the physical mechanism underlying this process [1].

Numerical simulation has clearly demonstrated that the generation of electron bunches by a laser pulse is a consequence of multithread motion of the electron component of plasma. Such a motion can be presented in the form of mutually intersecting trajectories of plasma electrons, each electron being initially a plasma oscillator excited by laser pulse, which executes free oscillations around the initial position, which this oscillator occupied prior to the action of the laser pulse. The necessary condition for oscillator trajectory intersection is an excess of their total energy E_{os} over the threshold value $E_{os\ th} = mc^2[1 - (V_{gr}/c)^2]^{0.5}$, determined by the group velocity of the laser pulse V_{gr}.

Intersection of the trajectory of the plasma oscillator with that of the neighbouring oscillator results in self-injection of this electron into the wake wave produced by the laser pulse, which is the physical mechanism of generating an electron bunch. However, not all electrons after self-injection are captured in the bunch produced, because the critical item is the position of the point of electron self-injection in the wake wave. The electrons whose self-injection point fits into the accelerating phase domain of the wake wave are kept in the generated bunch. The electrons, which perform self-injection near the boundary of the accelerating phase of the wake field, are only partially captured by the wake wave. Those can be captured only after the plasma background electrons, not participating in the process of electron self-injection, start intensively flow through the bunch of captured electrons.

For determining the parameters of the electron bunch generated by a laser pulse, we have analytically obtained the trajectory for the electron-leader, i.e., for the first electron injected into a wake wave, which resides in the head of the bunch. It was shown that at the formation stage of the electron bunch when background plasma electrons are self-injected into the wake wave, the duration of the electron bunch and the spread of the electron energy in the bunch are determined by the trajectory of the electron-leader and by variation of its energy along this trajectory.

Simple formulae are obtained for estimating the duration of the electron bunch and the energy spread of bunch electrons after the process of the bunch formation terminates. It is shown that characteristics of the generated electron bunch are determined by the group velocity of the laser pulse and by the energy of plasma oscillators excited by the pulse. It was found that the electron bunch duration may be ∼100 as at the relative spread of the electron energy less than 10%.

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Large beam effect in structuring of Si surface with ultrashort laser

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Nowadays, silicon is one of the most widely used material in nanostructure research and in technological, high precision applications including fine chemical industry and semiconductor electronics. However, the process of nanostructure is not fully explained and to control the speed of the process is still a challenge. Here, we report large beam effect in silicon nanostructure in fs and ps range. Considering laser-surface interaction parameters and experimental results, the main influence of the large beam area can be seen in increasing the interaction of surface scattered waves that will be discussed in this talk.

Dispersion law and damping of electronic high-frequency waves in plasma, formed by the tunnel ionization of atoms

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The dispersion laws and collisionless damping decrements of high-frequency electron waves in plasmas formed during the tunneling ionization of atoms by a powerful short laser pulse of circular and linear polarization are found. The effect of the wave propagation direction relative to the anisotropy axis of the photoelectron distribution on the frequency and damping decrement of plasma waves is investigated. With circular polarization of the radiation, it is established that propagation of weakly damped plasma waves with a linear dispersion law, whose frequency can exceed the Langmuir frequency by several times, is possible. In the case of linear polarization, the frequency of electron waves is comparable with the Langmuir.

A distinctive feature of plasmas formed during the ionization of gas atoms by a powerful short pulse of laser radiation is a velocity distribution different from the Maxwellian distribution of photoelectrons. For approximation, it is used that, in the tunneling ionization mode by a circularly polarized field, toroidal distribution of photoelectrons over velocities is realized, and in the case of linearly polarized radiation, bi–Maxwellian distribution of photoelectrons elongated along the polarization axis of radiation is formed. The dispersion laws and collisionless damping decrements of high-frequency electron waves obtained in this work demonstrate a highly anisotropic dependence on the direction of propagation of such waves relative to the anisotropy axis of the corresponding photoelectron distribution. It is found that for directions of propagation of plasma waves along which the average kinetic energy of photoelectrons is maximum, a strong dispersion and the largest damping decrement are observed.

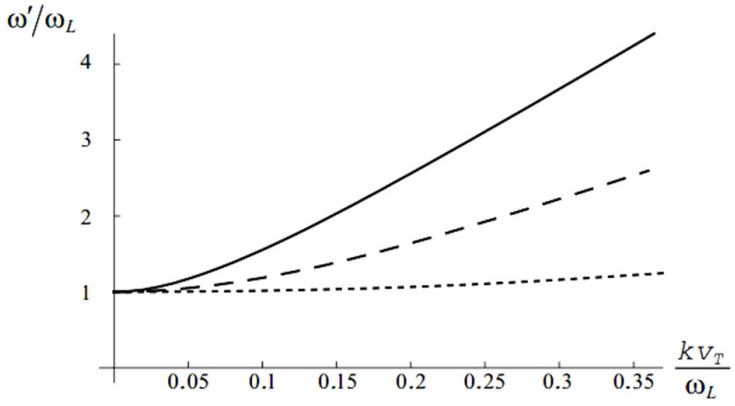

Figure 1: The dimensionless frequency and collisionless damping decrement of electron plasma waves in the case of circular polarization of radiation propagating at angles $\theta_k = 0$ - dotted line, $\theta_k = \pi/3$ - dashed line, $\theta_k = \pi/2$ - solid line.

In the case of circular polarization of ionizing radiation, it is shown that for a given wave number as θ_k increases, the angle between the wave vector $\bar{k}$ and the anisotropy axis, the frequency and damping decrement of quasilongitudinal waves increase substantially,

reaching its maximum values at $\theta_k = \pi/2$. The frequency of weakly damped plasma waves propagating at not small angles to the axis of anisotropy, when $k \sin\theta_k \gg \omega_{Le}/v_E$, substantially exceeds the Langmuir frequency of the electrons and is proportional to the wave number. The dispersion and damping of waves propagating strictly along the anisotropy axis are the same as in the Maxwellian plasma.

A strong dependence on the direction of the wave vector dispersion and collisionless damping of high-frequency plasma waves is also observed in a plasma obtained with linearly polarized ionizing radiation. In such a plasma, the average energy of the electrons along the axis of anisotropy, which coincides with the direction of polarization of the ionizing radiation field, is much greater than in the plane orthogonal to this axis. This is the reason why the frequency of weakly damped quasipotential plasma waves propagating at small angles to the axis of anisotropy is somewhat larger than for large θ_k. The corresponding differences in the wave frequencies from the Langmuir frequency are determined by small, quadratic in k dispersion corrections, which are proportional to the effective temperatures of photoelectrons along and across the anisotropy axis.

Femtosecond relaxation dynamics of hot electrons in high temperature cuprate superconductors

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After the interaction between a superconductor and a short laser pulse the hot electron ensemble relaxes to equilibrium state via energy exchange with bosonic field of both electronic and lattice origin, characterized by different relaxation dynamics on the femtosecond timescale. The large value of superconducting gap in cuprates permits create a transient non-thermal population by ultrashort and very intense THz and far-infrared (FIR) pulses. (As promising techniques for such experiments there can be considered sources of THz waves and FIR radiation, tunable with magnetic field, presented in [1]). The interaction of hot electrons with bosonic excitations can be described by some bosonic function, depending on the density of states of the excitations and the strength of their mutual coupling. The character of dielectric function in this case is determined by relation of the frequency-dependent dissipation of the Drude optical conductivity and a contribution of Lorentz oscillator(s) for interband (charge transfer (CT)) optical transition(s) (in the visible region for cuprates), which procedure, in principle, is well known (see, e.g. [1]). Available data on non-equilibrium optical spectroscopy in cuprates demonstrate that on the fast timescale ($\ll$ 100 fs) (faster than the electron-phonon thermalization) hot electrons are already thermalized with some bosonic excitations of electronic origin. The detailed analysis of time- and frequency-resolved pump-probe optical spectroscopy (reflectivity) data (wavelet analysis) allows in principle to distinguish electronic and phononic contributions. In present work, it is demonstrated that in cuprates it is necessary to take into account electronic excitations, corresponding to the in-plane CT-transition (CT-excitons). Such a picture corresponds to the excitonic mechanism of high-Tc superconductivity, proposed by V.L. Ginzburg well before (for details, see [2]). The parameters of in-plane CT-excitons in stripe structure, formed in conducting planes of cuprates due to magnetic (AF SDW) phase transition, are exactly within the range of optimal parameters for "Ginzburg sandwich" (nanoheterostructure: insulator-metal-insulator, in planar geometry). The presence of both electronic (excitonic) and lattice contributions in relaxation process is indicative to combined exciton-phonon mechanism of high-Tc superconductivity in cuprates [2].

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Wavelength scaling of deposited energy density under femtosecond microstructuring in bulk fused silica

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Femtosecond microprocessing is of particular interest for a number of areas in science and technology ranging from precise cell surgery to industrial fabrication of opto-electronic devices [1-7]. The dependence of deposited energy density (DED) on laser parameters provides unique information about the breakdown initiation and dominant mechanisms. Detailed knowledge of the parameter dependence of DED is essential for creation of controlled microstructures with predictable morphology at micro-and nano-scale.

Here we present a wavelength and energy dependence of the DED and the laser-induced damage threshold under a single femtosecond laser pulse processing in bulk fused silica. In the experiment, a pump-probe geometry was used. Fundamental radiation of Cr:Forsterite laser (1.24 μm), its second harmonic (0.62 μm) and signal wave of a home-built optical parametric amplifier (1.6—2 μm) were used as a pump. An aspheric lens with numerical aperture of 0.5 focused radiation in fused silica sample mounted on the 3D translation stage. The stage was moved continuously with laser repetition rate in order to avoid accumulation effects. The probe beam (1.24 μm) was focused by the same lens and generates the third harmonic (TH) on the microplasma produced by the pump beam. Additional lens mounted on the 3D translation stage was adjusted for spatial overlap of the beams. Translation of this lens allows 3D reconstruction of plasma-affected zone from the measured TH signal. To determine the DED, the nonlinear transmission of the pump beam was also measured. To reveal the relative contribution of the field-induced ionization and the impact ionization we employ a numerical model based on the Keldysh photo-ionization theory and accounting for the impact ionization by means of the multiple rate equations approach [5,8]. The model was used to simulate the evolution of the energy density gained by the electron subsystem during the laser pulse at various laser pulse peak intensities and wavelengths. We consider the laser-induced damage threshold being reached when the energy density in the electron subsystem at the end of the laser pulse reaches the threshold of a modification formation in fused silica.

We measured the evolution of the DED and the laser induced damage threshold at various incident wavelengths. The study reveals experimentally and numerically that in the visible range (0.62 μm) free electrons are produced by the multiphoton ionization followed by avalanche ionization, whereas in the mid-IR (2 μm) the dominant mechanism is tunnel ionization. We also found that the maximum energy deposition is achieved for short-wavelength laser pulses (0.62 μm), which is associated with a better spatial localization and a higher-density microplasma. At high incident energies, the DED saturates, and even slow decreases, remaining above the threshold of a modification formation (about 2 kJ/cm^3 for fused silica). On the other hand, the "plateau" region is reached faster for long-wavelength radiation (fig.1).

The simulations were performed on resources at Chalmers Centre for Computational Science and Engineering (C3SE) provided by the Swedish National Infrastructure for Computing (SNIC) project № SNIC2018-4-12.The experimental research has been supported

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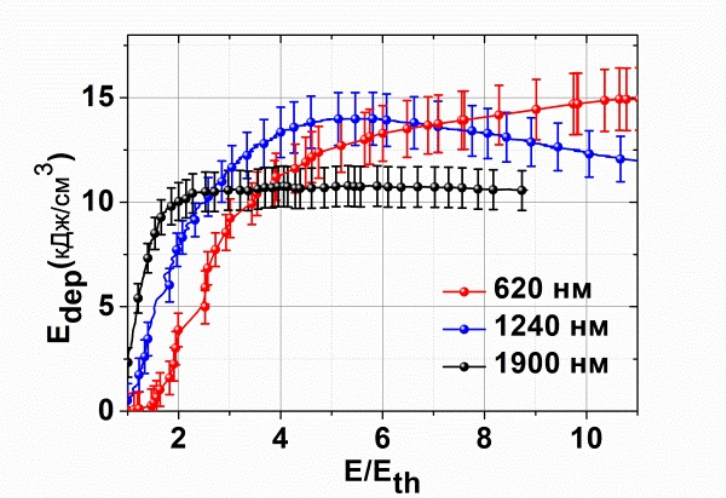

Figure 1: Energy and wavelength dependence of deposited energy density (E_{th} indicates the threshold energy of plasma formation E_{th}= 220 nJ for 0.62 μm, 0.6 μJ for 1.24 μm, 0.5 μJ for 1.9 μm).

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Transport and optical properties of noble metals at two-temperature state

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Two-temperature hydrodynamics and molecular dynamics simulations are the common methods used for modeling of surface nanomodification by a femtosecond laser irradiation. At the level of implementation of these methods, we should provide data about electronic conductivity and electron-phonon coupling. These characteristics of two-temperature state may be determined on the base of experimental data subjected to a processing by well-known models such as Drude model [1,2] or modified two-temperature hydrodynamics [3]. Therefore, an interpretation of such results depends on applicability and accuracy of the processing methods. In the case of pump-probe experiments [1,2], we can find out a change of optical properties of the metal surface at the stage of electron-ion relaxation where electrons are heated up to several eV. In the recent paper [2], measurements of dielectric permittivity of some metals in two-temperature state demonstrate significant changes in refraction and reflective coefficients. Interpretation of these data can be useful to check the data about effective frequencies of electron-electron and electron-ion collisions. In this work, we carried out calculations of transport and optic properties of noble metals using the Kubo-Greenwood approach and also two-parabolic approximation in conjunction with Drude model. Both results are compared with the experimental data given in the work [2]. The work was supported by Russian Foundation for Basic Researches (Grant № 16-20-00864).

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Role of surface plasmons in laser-induced THz generation from metals

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The talk is devoted to the review and theoretical interpretation of the last experimental results in laser-induced THz generation from metal structures. The special attention will be paid to the metal grating where resonant excitation of surface plasmons by the incident laser pulse is possible. Despite not a record efficiency level, study of optical-to-THz conversion on metal surface are relevant because of future applications in structural diagnostics and due to the fundamental interest in transformation mechanisms. Now there is no clear microscopic theory which is able to interpret the process of THz generation by metal gratings; in particular, the role of surface plasmons is not understood.

The first experimental measurements of THz signal from metal grating were performed more than 10 years ago. In general, the strong influence of surface plasmon excitation on the THz generation was experimentally proved for different experimental conditions. In the typical case a smooth resonant dependence of THz signal energy on the incidence angle is observed, where the resonant angle corresponds to the synchronism between the surface plasmon (on the optical frequency) and one of the diffraction waves. In the same time, one of the most complicated experimental features is non-quadratic dependence of the THz radiation energy on the optical fluence.

In the talk several possible microscopic mechanisms of plasmon-enhanced THz generation will be discussed. The first one is based on the emission and acceleration of electrons above the surface, the second one – on the ponderomotive action of the plasmon field inside the metal, the third one takes into account thermal effects in the electronic gas. We present some analytical results allowing to calculate the amplitude of the surface plasmon in the case of finite absorption and finite laser pulse length. This gives an opportunity to estimate the contributions of different mechanisms to the formation of THz response of metal gratings.

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Nanograting structures in transparent dielectric at the nonlinear stage of plasma-resonance instability

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Periodic structuring phenomena in the electromagnetic-wave-induced discharge processes were studied at different times as applied to various ionizable media, wave frequency bands, and manifestations of different types of ionization-field instability. Much recent attention has been focused on the sub-wave periodic plasma-field structures, that develops during fs laser-pulse-induced breakdown at the surface or within the volume of the solid transparent dielectric (in the main, the fused silica) and are considered as responsible for the volume nanograting formation in this material by the series of repeated pulses. The physical mechanisms determining the formation of these structures are not revealed completely and remain the subject of discussion. One of such possible mechanisms is the development of the ionization-field instability [1,2]. In this report, based on a simple initial-time model, we have present the results of numerical simulation for the spatio-temporal evolution of femtosecond laser-pulse-induced breakdown in transparent dielectric (fused silica) at the nonlinear stage of one kind of this instability, namely, so-called plasma-resonance one. The latter develops from very small seed perturbations of the medium permittivity and results in, because of the strong mutual enhancement of the electric field and plasma density perturbations in the plasma resonance region, the formation of the subwavelength periodic plasma-field structure consisting of the overcritical plasma layers perpendicular to the laser polarization. The calculation of the time-course and spatial profiles of the plasma density, field amplitude and energy deposition density in the medium during one breakdown pulse has allowed to establish the main possible scenarios of the process considered and to found the laser intensity and pulse duration ranges where this process can underlie the nanograting modification of the medium by repeated pulses.

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Nonlinear absorption of femtosecond light pulses under conditions of multiphoton resonances in bulk crystals and nanostructures

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A theoretical analysis has been performed of the nonlinear response of bulk materials and heterostructures with quantum wells, wires, and dots to ultrashort light pulses with a duration smaller than the time of intraband (intrasubband) relaxation of the electron's or hole's momentum. Analytical formulas are obtained for dynamic nonlinear polarizabilities, which control the energy absorbed from the ultrashort light pulse by structures of differing dimensionality under conditions of n–photon resonance on transitions between the discrete states or between the sublevels (subbands of size quantization). The dependences of the absorbed energy upon detuning of multiphoton resonances are obtained for bulk crystals and nanostructures. In a similar way, the dependences of the absorbed light energy on the ultrashort pulse duration are obtained under condition of fixed pulse energy. It is shown that, in the case of zero-dimensional objects (quantum dots and impurity centers) these dependences essentially differ from those taking place for relatively long pulses.

The interaction of two successive fs light pulses with solids and low–dimensional structures in the pump-probe spectroscopy mode was also considered. The dependences of the energy absorbed from the probe pulse on the two-photon resonance detuning have been obtained. Similarly, the dependences of the light energy absorbed from the probe pulse on the time delay between the pulses have also been obtained. We have also derived expressions for the power absorbed from the probe pulse depending on the time of interaction between the sample and the pulses. It has been shown that there is a significant difference in the behavior of the absorbed power for positive and negative time delays between the pulses.

A theory of the transient absorption of femtosecond light pulses at a two–photon-one–photon double resonance on adjacent interband transitions in crystals has been developed. Approximate expressions for the reactive component of the nonlinear polarization of the quasi–three–level system, P_s, which determines the absorbed power in the medium, have been obtained, and the dependences of P_s on the light intensity and detunings of resonances have been calculated. The dependences of the energy that is absorbed in a crystal exposed to the action of a femtosecond light pulse on the radiation intensity at a fixed pulse duration and on the pulse duration at a fixed pulse energy have been determined.

We have developed a theory of the transient absorption of femtosecond light pulses by free electrons in the course of intraband transitions involving the participation of phonons. General expressions have been obtained for the probability of transition of an electron from the bottom of the conduction band to a highly excited state due to the absorption of several photons and either the absorption or emission of a phonon. For arbitrary integers n, we have calculated probabilities W_n of n–photon intraband transitions with the participation of longitudinal acoustic and longitudinal optical phonons. Dependences of W_n on the duration of femtosecond pulses have been obtained. These dependences differ from those that are observed in the case of quasi-steady electromagnetic radiation.

New approach to the problem of THz generation in high-intensity laser field

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New approach to construct the source of radiation in THz frequency band is discussed. It is based on formation of the THz spontaneous background by the strong-field ionization of atoms in two color femtosecond laser pulse. This background further can be significantly enhanced in nonequilibrium plasma channel formed by powerful short laser pulse in heavy rare gases with Ramsauer minimum in the transport cross section.

Relaxation dynamics of nonequilibrium electrons in laser-excited solids

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When an ultrashort laser pulse of visible light is absorbed by a solid, mainly the electrons in the material are excited. In metals, free electrons in the conduction band can directly absorb photons. In semiconductors and dielectrics, on the other hand, a band gap has to be overcome first, as almost no free electrons are present at room temperature in the unexcited material. Due to this excitation, the electronic system, or the so–called electron–hole plasma, is in a nonequilibrium state. A sequence of different relaxation processes transfers the material into a new equilibrium. Depending on the interaction associated with the particular relaxation process, it occurs on a characteristic timescale.

On the basis of complete Boltzmann–type collision integrals, we calculate the transient distribution functions of electrons and phonons in different materials. We consider electron–electron interaction, different ionization processes, as well as electron–phonon coupling. By that we trace the relaxation cascade of nonequilibrium electrons after ultrafast heating.

Distinct material properties enter through the density of states of the electrons in the conduction band. We study in particular noble metals, dielectrics and ferromagnets. In noble metals and ferromagnets, d–electrons play an important role, whereas in dielectrics two separated bands govern the dynamics and the ionization state may differ from. We show, that the electron distributions deviate from Fermi distributions for timescales up to a few picoseconds. While the initial thermalization within one band has an intrinsic timescale of typically only a few tens of femtoseconds, nonequilibrium occupations of the different bands as well as continous electron–phonon coupling drive the conduction electrons out of equilibrium for much longer times [1, 2].

We present in detail the mutual influence of different interaction and relaxation processes.

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Layer-by-layer modification of thin-film metal-semiconductor multilayers with ultrashort laser pulses

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The surface modifications in a multilayer thin-film structure (50-nm alternating layers of Si and Al) induced by a single Gaussian-shaped femtosecond laser pulse (350 fs, 1028 nm) in the air are investigated by means of atomic-force microscopy (AFM), scanning electron microscopy (SEM), and optical microscopy (OM). Depending on the laser fluence, various modifications of nanometer-scale metal and semiconductor layers, including localized formation of silicon/aluminum nanofoams and layer–by–layer removal, are found. While the nanofoams with cell sizes in the range of tens to hundreds of nanometers are produced only in the two top layers, layer–by–layer removal is observed for the four top layers under single pulse irradiation. The 50-nm films of the multilayer structure are found to be separated at their interfaces, resulting in a selective removal of several top layers (up to 4) in the form of step-like (concentric) craters. The observed phenomenon is associated with a thermo-mechanical ablation mechanism that results in splitting off at film–film interface, where the adhesion force is less than the bulk strength of the used materials, revealing linear dependence of threshold fluences on the film thickness.

In summary, on the basis of the carried out experiments, one can suggest a qualitative picture of the observed phenomena. The laser energy is absorbed by the two top layers that results in its heating and melting at a time scale of ~ 1 ps. Due to isochoric heating a rarefaction wave is produced that penetrates deep into the sample simultaneously with the thermal wave and causes rupture at the boundary of layers, where the strength is weaker. The presence of the enormously high and melted corrugated nanostructures (rims) at the edge of all the removed layers might indicate the simultaneous influence of acoustic and thermal waves at the studied spatial scale of 200 nm. For the time being, additional experiments and a rigorous theoretical model that takes into consideration singularities of thermal conduction in nanoscale layers, interlayer adhesion and Kapitza resistance are necessary to elucidate the observed formation of nanofoam and layer–by–layer removal of thin multilayered films excited by a single ultrashort laser pulse.

Laser induced breakdown spectroscopy principles and ultrashort pulses' effects

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Laser matter interaction, especially using ultrashort laser pulses, involves many linear and nonlinear mechanisms which determine sample mass removal and relative plasma emitting features to be detected. In this presentation, an overview on the main processes induced after absorption of laser pulses by the sample, such as electronic excitation and ionization, non thermal melting and electron lattice heating will be discussed together with successive formation of the emitting plasma, its evolution in both ns and fs regimes as well as, for the latter, nanoparticles formation. Peculiarities on low thermal heating and lateral damage of the sample after incidence of ultrashort laser pulses have determined their success for surface treatments of materials as well as for Laser Induced Breakdown Spectroscopy (LIBS) applications. It has been demonstrated, because of a low emitting continuum contribution, that for these laser pulse durations it is feasible to perform LIBS measurements even without the employment of gated detectors. This advantage, nevertheless, does not take into account the accomplishment of the fundamental main requirements for LIBS compositional analysis of materials. In this context, an introduction on low plasma emission intensity, mechanisms and experimental observations regarding excitation and deexcitation processes due to electronic inelastic collisions, plasma inhomogeneity, Local Thermodynamic Equilibrium (LTE) or partial LTE (p-LTE) and their lack or fulfillment, is going to be drawn. In order to overcome drawbacks and enhance the main benefits offered by ultrashort laser pulses for LIBS applications, some strategies will be illustrated. Experimental evidences of successful recent LIBS applications performed by ultrashort laser pulses together with their common trends and remarks on possible future growths will be discussed.

Self-induced transparency of intense few-cycle terahertz pulses in n-doped silicon

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Intense few cycle terahertz pulses may exhibit complex non-linear behavior under interaction with heavily n-doped Silicon. We show that a fast increase in the transmission of the 700 fs electromagnetic pulse (central frequency ∼1.5 THz) through the 245–μm thick n-doped silicon sample with carrier concentration of 9×10^{16} cm^{-3} (low field transmission of ∼0.02%) saturates at the field strength of ∼5 MV/cm^{-1} at ∼8% and then drops twofold at ∼20 MV/cm^{-1}. Electro-optical sampling measurements reveal formation of a single cycle terahertz pulse at highest field strengths due to an efficient cut-off of all field oscillations except for the first one by a thin layer ionized by this oscillation.

Volume nanostructuring with spatio-temporally sculpted laser pulses

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Nowadays micro/nano–technologies are dependent on the development of processing tools able to structure materials in two– and three–dimensions with utmost precision. In this respect ultrafast lasers hold great promise for taking up ambitious processing challenges. We discuss the potential of volume micro and nanostructuring to the field of photonics, notably for the development of optical functions and devices based on laser-induced 3D refractive index engineering where nanoscale accuracy can deliver high levels of performance. Controlling energy delivery in space and time can achieve record precision at the 100 nm scale. We discuss a range of physical mechanisms leading to the fabrication of features below diffraction limit. The potential of volume hybrid micro-nanoscale structures is outlined by a range of applications examples in sensing, spectroscopy and telecom.

Role of continuous quasi-stable states in high order harmonic generation. Resonance-induced modification of harmonic spectrum and phase-locking

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High harmonic generation (HHG) via interaction of intense laser radiation with matter provides a unique source of coherent collimated XUV femto- and attosecond pulses. Recently much attention has been paid to the role of resonances in HHG in gases and plasma plumes. It was shown that when the high-harmonic frequency is close to the transition to an excited quasi-stable state of the generating particle, the harmonic can be much more intense than the nonresonant ones.

We suggested a HHG theory [1] which generalizes the strong-field approximation to the resonant case when the harmonic frequency is close to that of the transition from the ground state to an autoionizing state (AIS) of the generating system. To describe the resonant HHG we add a quasi-stable state to the system of the ground and free continuum states considered in the nonresonant case in Ref. [2]. The interference of the nonresonant and the resonant terms leads to the Fano-like factor in the harmonic line shape. To check our analytical predictions, we develop a numerical approach based on the time-dependent Schrödinger equation solution for a model potential.

Our theory confirms the four-step resonant HHG model [3], where the first two steps are the same as in the three-step model [4], but instead of the last step (radiative recombination from continuum to the ground state) the free electron is trapped by the parent ion, so that the system (parent ion + electron) lands in the quasi-stable state, and then it relaxes to the ground state emitting XUV.

The results of the calculations of the harmonic intensities are in good quantitative agreement with the experiments on HHG in plasma plumes showing efficiency growth up to two orders of magnitude when the harmonic is resonant with the transition frequency. The calculations reproduce well the essential difference in HHG efficiency for different ions.

Our simulations predict phase-locking of the resonantly-enhanced harmonics and strong influence of the resonance at the harmonic phases. This conclusion was confirmed by experiments using RABBIT technique [5]. The measured harmonic phases are in good agreement with our numerical calculation [5], and reasonably agree with our theoretical predictions.

The phase-locking of the resonant harmonics and relatively high conversion efficiency make them interesting for the attosecond pulse generation. The attosecond duration of the XUV pulse assumes broadband resonant enhancement; such enhancement was observed in Xe for XUV near 100 eV in the spectral region of about 20 eV [6]. We study theoretically [7] the effect of giant resonance in Xe on the phase difference between consecutive resonantly enhanced harmonics and calculate the duration of the attosecond pulses produced by these harmonics. For certain conditions, resonantly induced dephasing compensates the phase difference which is intrinsic for nonresonant harmonics (the so-called attochirp). We find these conditions analytically and compare them with numerical results. This harmonic synchronization allows attosecond-pulse shortening in conjunction with the resonance-induced intensity increase of more than an order of magnitude. The

latter enhancement relaxes the requirements for the UV filtering needed for attosecond-pulse production. Using a two-colour driving field allows a further increase of the intensity. In particular, a caustic-like feature in the harmonic spectrum leads to a generation efficiency growth of up to two orders of magnitude, which is, however, accompanied by an elongation of the XUV pulse.

Studies of the resonant HHG using finely-tuned laser wavelength [8] show that resonant generation involves not only the quasi-stable AIS, but also a harmonic generation from dressed-AIS that appears as two coherent satellite harmonics at frequencies $\pm 2\omega_{las}$ from the resonant. Thus, the resonant HHG allows not just enhancement of the generation efficiency but also observation of the new phenomena like dressed-AIS. Moreover, measuring of the resonant harmonic phase can provide access to the new spectroscopic information dealing with the *phase* of the dipole matrix element of the ground-AI state transition.

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High-frequency longitudinal waves in plasma generated by multiphoton ionization of gas atoms by a short laser pulse

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Plasma with non-equilibrium photoelectron velocity distribution is formed at the ionization of gas atoms by the field of short pulses of intense laser radiation. The distinction of the photoelectron distribution from the Maxwellian one can lead to the appearance of a number of new physical phenomena and qualitative change in the properties of already known processes. In this case, the explicit form of the formed photoelectron distribution plays an important role. Under the conditions when laser pulses have relatively high intensity, the ionization of atoms is associated with the tunneling effect and the formed anisotropic photoelectron distribution function leads to the appearance of new optical properties of the photoionized plasma, new properties of surface waves at the boundary of such a plasma, the possibility of developing aperiodic instabilities and amplifying of low-frequency radiation [1,2]. In the case of plasma formation in the multiphoton ionization regime, which is realized at low intensities of ionizing radiation, the formed photoelectron distribution relatively quickly becomes isotropic and the problem of instabilities associated with the anisotropy becomes insignificant. At the same time, the photoelectron energy distribution that arises in the case of multiphoton ionization of atoms is strongly nonequilibrium and consists of a set of relatively narrow energy peaks corresponding to the absorption of definite quantity of photons, which also leads to the appearance of unusual properties of the formed plasma.

In this communication for weakly ionized plasma formed at multiphoton ionization of xenon atoms it is shown that the properties of high-frequency electron plasma waves in such plasma differ qualitatively from properties of Langmuir waves which are the analogs to those waves in Maxwellian plasma. When photoelectron distribution is mainly represented by electrons appearing at three-photon ionization of atoms, it is shown that potential high-frequency waves with frequency greater than electron plasma frequency exist in photoionized plasma. Dispersion law of these waves is close to linear in the range of not very small wave numbers and their phase and group velocities are determined by the average velocity of photoelectrons. The collisionless damping of these waves, in turn, is exponential low in wide range of wave numbers, but sharply increases when wave number comes closer to the value equal to the ratio of plasma frequency to the velocity determining the spread of photoelectrons over energy.

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Peculiarities of interaction of doughnut-shaped laser pulses with transparent materials

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The interaction of femtosecond laser pulses of toroidal spatial shape with the bulk glass has been studied numerically in the frames on non-linear Maxwell's equations [1,2]. It has been found that, compared to Gaussian laser pulses (GLP), doughnut-shaped laser pulses (DSLP) allow to achieve much more localized laser energy absorption. The threshold for glass modification in the DSLP cases can be several times smaller than for the GLP at the same focusing conditions while, at the same beam energy, the peak absorbed energy density is more than the order of magnitude higher for DSLP. A typical distribution of the absorbed laser energy density in the case of DSLP has a form of a cylinder with very hot walls and a relatively cold core. Such distribution must result in the emission of shock waves convergent to the axis of the cylindrical region, thus providing extremely large compressive stresses in this region. These DSLP features can find applications for enhancing precision of laser processing of materials at lower laser energy, generation of extreme states of matter, and in bio-medicine.

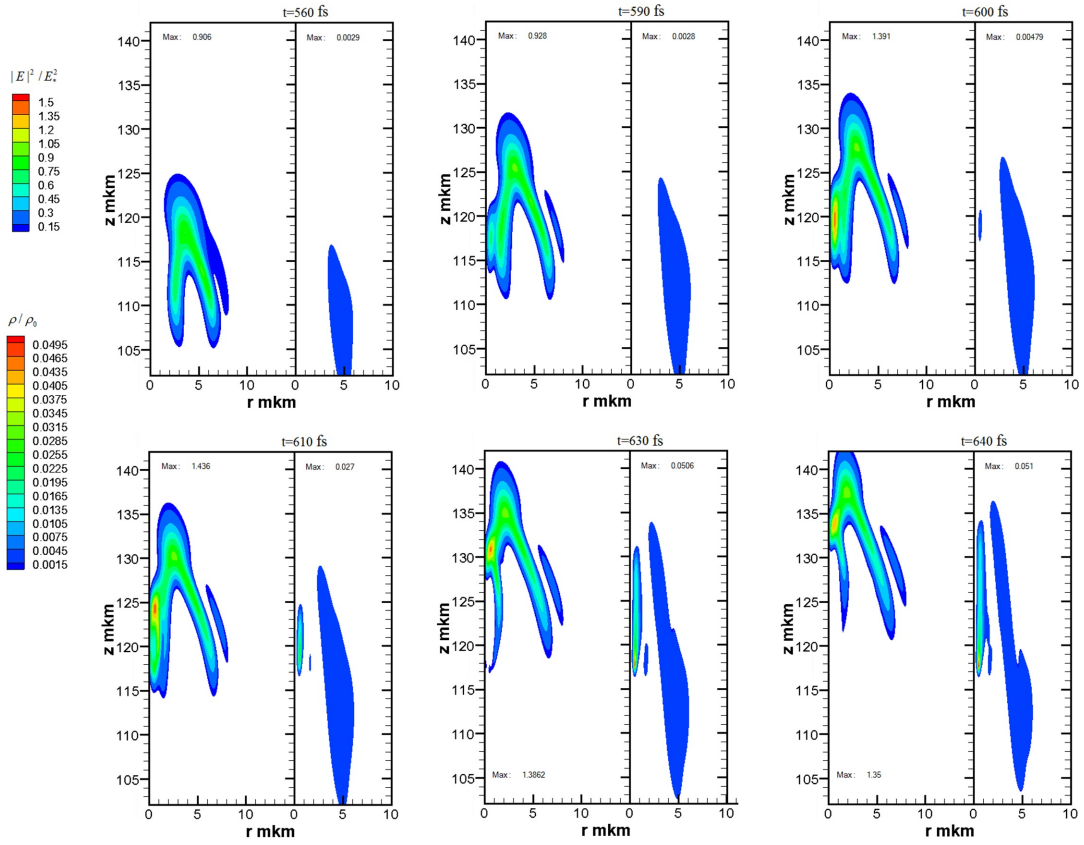

Figure 1: Distributions of laser intensity (E^2, left panels) and free electron density (ρ, right panels) at different time moments upon propagation of DSLP in fused silica. Pulse duration is 45 fs; numerical aperture of focusing lens is 0.25.

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Section 3: Ultrafast laser nanofabrication and nanophotonics

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Scope
Ultrafast nanostructured light + nanostructured matter
Ultrafast nanophotonics
Femtosecond-laser nanofabrication

The effect of delay time between the pulses of different frequences in surface nanopatterning by two-colored femtosecond pulses

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We consider femtosecond laser nanostructuring of the material surface by means of a colloidal particle lens array. Here, the monolayer of dielectric micro- or nanospheres placed on the surface acts as an array of near-field lenses that focus the laser radiation into the multitude of distinct spots, allowing the formation of many structures in a single stage. Previously, it was shown that conversion of a small part of the energy of the fundamental frequency (FF) femtosecond beam into the second harmonic (SH) is an efficient way to decrease the modification threshold and to increase the surface density of obtained nanostructures.In the present communication, we investigate the effect of delay time between the FF and SH pulses on the efficiency of the nanostructuring. The result depends on the position of the edge of the absorption band of the substrate with respect to the photon energy of the third harmonic (TH). If the TH photon energy is larger than the band gap, then the minimal modification threshold corresponds to the zero delay time between the pulses because the two–photon absorption FF+SH dominates the material alteration process. When the delay time is larger than the pulse duration that is the two-photon absorption is excluded, then the threshold is smaller when the SH pulse comes first. It can be explained by the different role of the FF and SH in multiphoton ionization that is responsible for the transparent materials alteration by femtosecond pulses. We also consider the two-pulse (FF and SH) interactions with the substrates with the band gap larger the TH photon energy. When the TH photon energy is close to the band gap then the frequency chirp obtained by the both FF and SH femtosecond pulses influence the delay time dependence of the threshold. The results shed light onto the features of the interaction of two-colored femtosecond impulses with the transparent dielectrics. We discuss the advantages of employment of a colloidal particles lens array for the study of light-matter interaction in this case. The work is supported by RFBR (Grant № 16-02-00792).

Single-shot laser-induced formation of nanoparticles from thin silver films submerged in various liquids

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In this study, we show that Ag thin films (10 nm thickness) deposited on a glass substrate can be transformed into nanoparticles by laser-induced dewetting using a pulsed Nd:YAG laser operated at λ=532 nm. The film could be entirely dewetted by a single pulse, and required fluence was about 100 mJ/cm^2. The size distribution of produced particles depends on the properties of surrounding media: air or liquids with various viscosity, where exposure takes place, due to the nanoscale Rayleigh-Taylor instability. Pressure gradients developed in the evaporated fluid region when the thin film is rapidly melted inside a bulk fluid, produce strong forces causing the instability. The properties of fabricated nanostructures are investigated by scanning electron microscopy and optical spectroscopy. The absorption spectra of dewetted films were in well consistent with the surface plasmon resonance behaviors of metal nanoparticles. This process provides a facile and scalable method of forming metal nanoparticle arrays for plasmonic and other applications.

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Numerical modeling of electromagnetic response of Si nanosheets covered by plasmonic layers

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Not long ago received new silicon structure in the form of nanosheets is of interest for application in sensors, medicine and other fields. Preliminary structuring of silicon in such way allows to adjust the shape of deposited metal. Properties of such structures were further investigated by numerical simulation in comparison with experimental data. Comparison between electromagnetic response in visible and NIR range of silicon nanosheets with and without deposited gold has been made. It was shown that geometry of metal deposition and its amount strongly affect transmittance spectre of the structure. Spatial distribution of electromagnetic near fields inside such structures was also calculated.

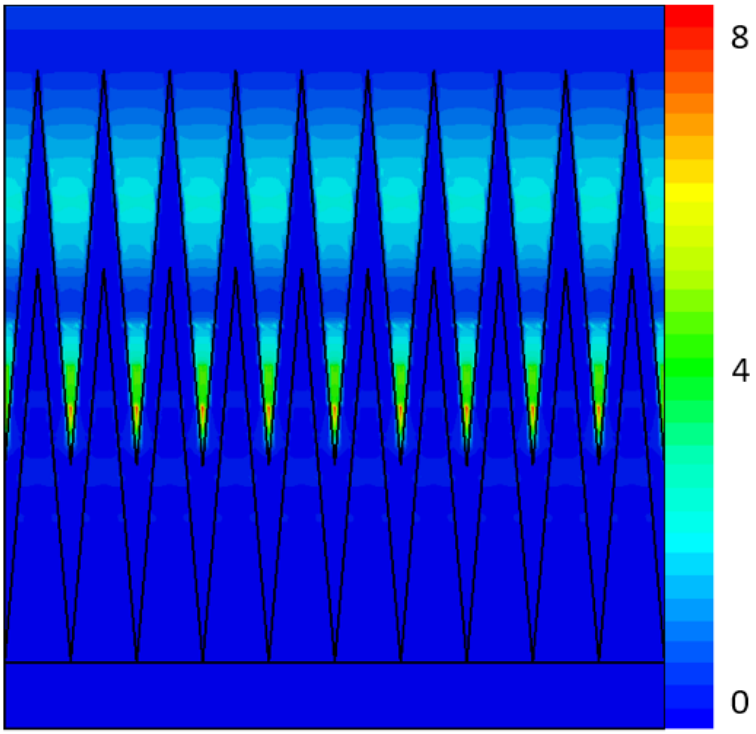

Figure 1: Spatial distribution of |E| in the case of silicon nanosheets covered by gold with a thickness of 300 nm. Color scale shown in |E| of incident plane wave (arb. units).

Processing dielectrics with controlled cracks from elliptical Bessel beams

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Ultrafast lasers have been a revolution in a number of areas of science and technology. In materials processing, the ultrafast pulse duration brought the ability to deposit energy into the free-electron gas before it couples to the lattice and therefore generate extremely steep thermodynamic gradients. A second benefit is the ultra-high peak power which allows for ionizing dielectrics, even from the bulk material. The processing of glass and sapphire is of particular importance for several technological applications to mass fabrication (consumer electronics, microelectronics, etc). In this field, nondiffracting Bessel beams have brought the ability of controlling energy deposition by a single laser pulse on a long propagation distance inside dielectrics. In particular, this has allowed avoiding nonlinear instabilities generated by the filamentation regime of Gaussian beams yielding inhomogeneous damage in materials [1].

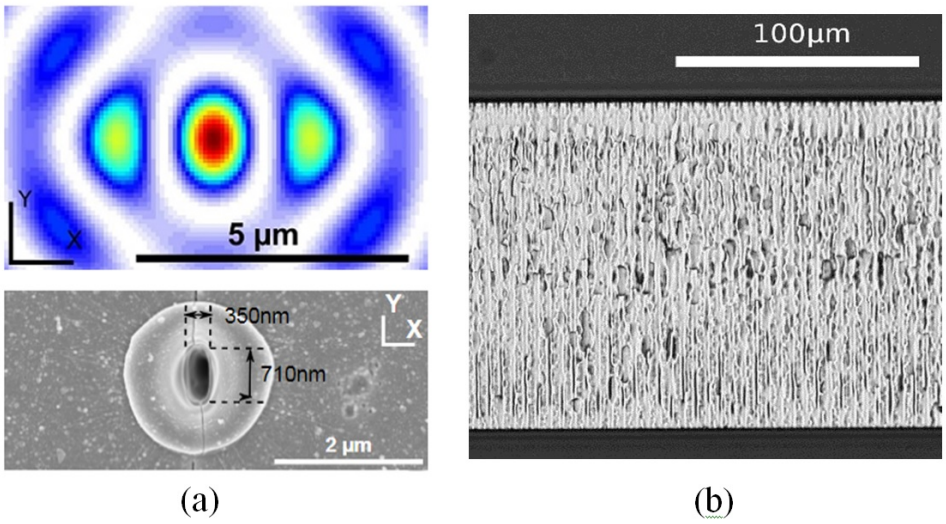

Figure 1: Figure 1 (a) (top) cross section of the elliptical Bessel beam developed. (bottom) Elliptical cross section of a nanochannel produced. (b) Side imaging of a sample cleaved after elliptical Bessel beam processing.

We report further advances in this field where additional benefit can be found in other beam shapes and pulse sequence. For applications to the so-called "stealth-dicing" technology, where material is cleaved after laser illumination, we have developed elliptical Bessel beams. We have demonstrated these beams remain also propagation-invariant and allow for generating nanochannels with elliptical cross section (Fig. 1(a)) [2]. We have demonstrated those allow for cleaving glass (Fig 1(b)) with enhanced reliability and with sub-μm accuracy [3].

In addition, we will discuss recent novel results where the illumination has been performed by double pulses with equal power. We show that, depending on the initial pulse energy, double femtosecond pulses enhance the ability of channel drilling in glasses. In particular, channels with larger diameters are produced [4].

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High-precision direct laser processing of plasmonic films by structured laser beam

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Direct laser structuring of thin metal (plasmonic) films is a versatile, high-performance method for the fabrication a variety plasmonic elements. Single tightly focused short and ultrashort laser pulses are used to fabricate different types of nanostructures: microbumps, nanojets, nancrowns, micro – and nanoholes and their regular arrays. In this work, we used different optical devices, which generate complex intensity distribution of laser radiation. This method provides fabrication of plasmonic structures unusual forms. The size of elements depends on the focusing devices. High-NA focusing of shape-modulated laser beams provides single-shot printing of complex intensity patterns with significant increased focal depth.

Modification of Ti/Zr multilayer by femtosecond laser pulses

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Femtosecond laser texturing holds promise for the surface modification of materials, due to a wide application to all materials; the possibility of getting a wide variety of structures with micro- and nano-scaled features; and a fast, repeatable and contactless process. Laser processing is unique method, which allows production of active surface with formation of the desired oxide, creation of nano/micro textures and change wettability of the surface.

Due to excellent mechanical properties and moderate biocompatibility, Ti/Zr multilayer thin films, as novel nanolayered composites were deposited by ion sputtering on Si substrate. Subsequently, the Ti/Zr thin films were irradiated by femtosecond laser pulses in air to induce the following modifications: (i) mixing of components within the thin film structures, (ii) formation of ultrathin oxide layer at the irradiated surfaces, and (iii) structuring of the surface arrays in form of ripple structure. The main focus of this experimental study was examined different surface motives with nano- and micrometre features. For this purpose, the modifications of Ti/Zr multilayers were included formation of spots, lines and surfaces with different pulse energy, scanning speed i.e. number of pulses, z-distance. Laser-induced spots are composed of concentric circles, where the number of circles in individual spots is increased with increasing pulse energy. Maximum depth in the centre of spots and total roughness are gradually rising with pulse energy, but heights between ablated layers in these spots does not match with the thickness of layers, but these deviations are not significant. Lines and surfaces were scanned with different scanning rate, the conditions for formation of well-defined LSFL (low spatial frequency LIPSS) are determined. The periodic structures at high scanning rate (3 mm/s) are mainly formed on the multilayer thin film. However, the degree of ablation becomes higher at the lowest rate (0.5 mm/s) where the ripples mostly are formed on the Si substrate.

Heterochain thermostable polymers for the formation of optical elements

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The development of new materials and methods for the formation on their basis of various objects with high resolution is now one of the most urgent trends in modern science, especially in optics. In connection with the intensive development of high-power laser systems, there is a need for optical elements that can operate at high temperature and increased power density of laser radiation.

New promising methods for creating optical elements are additive technologies that use various transparent polymers [1]. However, the use of polymeric materials is very limited due to their decomposition under high temperature conditions and/or high power density of laser radiation passing through them.

For carrying out experiments on laser two-photon photocuring, a high-frequency ytterbium laser and a second harmonic generator from Avesta were used to obtain femtosecond laser pulses at a wavelength of 520 nm. Scanning of samples was carried out by a combination of precision translators produced by Aerotech and a two-mirror scanning head manufactured by Scanlab. This moving system allows you to achieve positioning accuracy of 50 nm and the highest performance available at the current time. To adjust the frequency of laser pulses, an acoustooptic modulator with a maximum switching frequency of 1 MHz is used. The system for moving the sample and modulating radiation is integrated into a single complex under the control of special software that allows processing according to a predetermined computer model. Different print resolution in experiments was achieved by using a set of focusing lenses with numerical apertures from 0.2 to 1.2.

The heterochain thermostable polymer - poly-N, N'- (m-phenylene) isophthalamide as a polymer photocurable material, and diacridamide 4,4'-diaminodiphenyloxide as a cross-linking agent was developed and synthesized in the Baikal Institute of Nature Management of the Siberian Branch of the Russian Academy of Sciences (BIP SO RAN). This polymer is unique in terms of preserving its properties under high temperature conditions. Preliminary studies showed its stability in the temperature range up to 400 degrees Celsius without significant degradation of its properties.

The paper shows the results of the development and application of new thermostable photocurable materials that retain their optical and mechanical properties at temperatures up to 400 degrees Celsius [3,4], which, in tandem with two-photon photopolymerization, can become a promising tool for the rapid and qualitative creation of optical elements for use in powerful laser systems. Their optical, mechanical properties are determined.

The formation of objects with high spatial resolution based on thermostable polymers preserving their optical and mechanical properties under elevated temperatures (up to 400 degrees Celsius) is shown for the first time, and the optical properties of the synthesized thermostable photocurable heterochain polymer compositions are determined.

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The dynamics of laser ablation thresholds of aluminum and steel during double-pulse femtosecond laser action

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Femtosecond laser processing of metals is used in various fields of science and technology. Some techniques of processing use pulse trains and double pulses. This introduces new physical phenomena [1, 2] that have to do with the change of the optical parameters of the material and require new in-depth studies of the physical processes during such treatment.

The aim of this work was to investigate ablation parameters (ablation thresholds, diameter and structure of the crater, the profile and depth of ablation tracks) of aluminum and steel during double-pulse femtosecond laser action depending on the thickness of the material and the delay between the pulses.

For targets we used four thin aluminum films (30, 60, 90 and 120 nm), a bulk aluminum plate and a bulk steel plate. Both plates were polished to be optical mirrors, the steel was AISI 304. Avesta-Project laser system with a wavelength of 800 nm, pulse duration of 100 fs, repetition rate of 10 Hz and a pulse energy of 1 mJ was used to carry out the experiment. The beam entered the optical scheme of a Michelson interferometer, where one arm was moved to provide the range of delays between pulses from 330 fs to 4 ns. After splitting both beams were converged $\sim$300 μm under the surface of the target to avoid filamentation. For photoacoustic measurements an ultrasound transducer with a band pass of $\sim$5 MHz and an oscilloscope Tektronix 3052C was used.

After the ablation procedure, the samples were studied by means of optical microscopy and profilometry. Ablation thresholds were determined; for bulk targets, the profiles of ablation tracks were obtained and investigated; for aluminum films, the dynamics of transmission during double-pulse femtosecond laser irradiation were also studied. Ablation threshold for aluminum targets show a similar dependence: ablation thresholds in double-pulse regime is higher than for monopulse and increases after $\sim$10 ps, reaching maximum in the range of delays from 60 to 100 ps, which we believe has to do with an expansion of material. Also, ablation thresholds (both for double pulses and monopulses) increased with an increase of the thickness of the target. Ablation thresholds for steel demonstrated another dependency: at the delays from 10 to 100 ps the threshold decreases, reaching the monopulse value at long delays. For both materials, the amplitude of the photoacoustic signal increases with a delay, while the transient time decreases, indicating the absorption of the second pulse in the expanding plume after the first pulse.

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Ultrafast laser-driven self-assembly and self-organization far from equilibrium

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We, humans, envy the level of complexity found in Nature, but we tend to regard its replication in human-made systems as an unattainable goal. This perception of un-unravelability of complexity is strongly biased by our dependence on, as well as the successes of, the deliberate linearization and suppression of noise that is pervasive in engineered systems. Can superior structures and functionalities, which are difficult or even impossible to achieve with linear and deterministic systems, be obtained by exploiting nonlinear dynamics far from equilibrium? Could control of complex systems, at times, be "simple"?

After giving a brief historical introduction to the complexity science, I will introduce ultrafast lasers as an excellent tool to control complex forms and functions by creating well-controlled spatio-temporal thermal gradients in far-from-equilibrium settings: Nonlinear interactions give rise to multiple fixed points in phase space during the dynamic evolution of the system. Each of these steady states correspond to a different pattern and their bifurcations. Amplified fluctuations, as a result of far-from-equilibrium dynamics, spontaneously drive transitions through bifurcations. Positive and negative feedback support exponential growth of perturbations, and their stabilization, respectively. This, in turn, allows control of great many degrees of freedom through the control of just a few. As concrete examples, I will demonstrate emergence of complex patterns and dynamic behavior from self-assembled polystyrene colloids (*Nature Commun.*, 2017). I will also argue that this is a "universal" dynamic self-assembly methodology by showcasing four different microorganisms, namely, *S. cerevisiae* (immotile, about $\sim$7 μm large, and elliptical in shape), *M. luteus* (spherical in shape, $\sim$500 nm in diameter), *E. coli* and *P. aerugosa* (motile, rod-like with dimensions of $\sim$1 μm x 2 μm), sub-5 nm particles and cancer cells (immotile, about $\sim$40 μm large). Finally, I will briefly show how we create self-organized nano and microstructures (*Nature Photon.*, 2013 and *Nature Photon.*, 2017) by invoking nonlinearities in the form of positive feedback between laser beam-induced changes in the material and material change-induced effects back on the laser beam.

Modelling of short laser pulse nanostructuring of metals in different media

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The possibility of materials surfaces restructuring on the nanoscale due to ultrashort laser pulses has recently found a number of practical applications in IT- and Bio- technologies. It was found experimentally that under spatial confinement due to a liquid layer atop the surface, one can achieve even finer and cleaner structures as compared to that in air or in vacuum. The physical mechanism of nanostructuring processes, however, is difficult to study experimentally since a lot of fast and non-equilibrium laser-induced processes remain beyond the attainability of the experimental measurements while the solid is in a transient state. The theoretical methods, on the other hand, working within a single computational approach, are frequently limited to the corresponding spatial and temporal scales. In this work we propose a combined atomistic-continuum (Molecular Dynamics based) approach, which was found as an efficient numerical tool in investigation of the nanostructuring mechanism of metals in super-large simulations. The combined model is applied to investigate the nanostructuring mechanism under conditions of vacuum ambient and in the regime of spatial confinement due to a thick water layer above the target. The simulation results are directly compared with the experimental data, generated on the same temporal and spatial scales, and analyzed. This allowed to extract the main mechanisms of nanostructuring process and the reasons for a higher quality of structures generated under water. It was demonstrated that depending on the applied fluence, the pulse shape, and the surrounding media one can achieve both the resulting surface and generated nanoparticles of specific character. Combined with the laser-controlled processes induced in materials, the ability of generation of surfaces with predesigned properties (topological, electrical, and optical) opens up a great potential for their utilization in the present for-front technologies.

Fabrication of hybrid Si-Au nanoparticles by nanosecond laser ablation

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In this paper, colloidal solutions of hybrid metal-dielectric silicon-gold nanoparticles were synthesized by nanosecond laser ablation in water from solid target of monocristal silicon wafer with thin gold film about 50 nm thinckness. The production of nanoparticles occurred at different scanning regimes: the laser radiation energy density (70, 100, 120 mJ/cm^2) and scanning speed were varied (50–750 mm/sec). Observation of hybrid nanoparticles occurred on a SEM, where an elemental analysis was made showing the fusion of gold and silicon, with the predominance of gold nanoparticles on the silicon surface (fig. 1a). Optical properties were studied in the solutions obtained by determining the extinction spectra of colloidal nanoparticles solutions using a spectrophotometer and calculating the extinction coefficient at wavelength 520 nm, were gold nanoparticles demonstrated plasmon resonance, from them to compare the yield of nanoparticles from the target under different scanning regimes (fig. 1b). At the inset SEM image hybrid Si-Au NP dried on fig. 1b it can be seen that an increase in the scanning rate leads to an increase in the dimensions of the generated nanoparticles. Thus, it is possible to influence the size and concentration of the resulting aqueous solutions of hybrid silicon-gold nanoparticles by varying the scanning modes of the laser system for their further use in various fields, for example, in the theatronics.

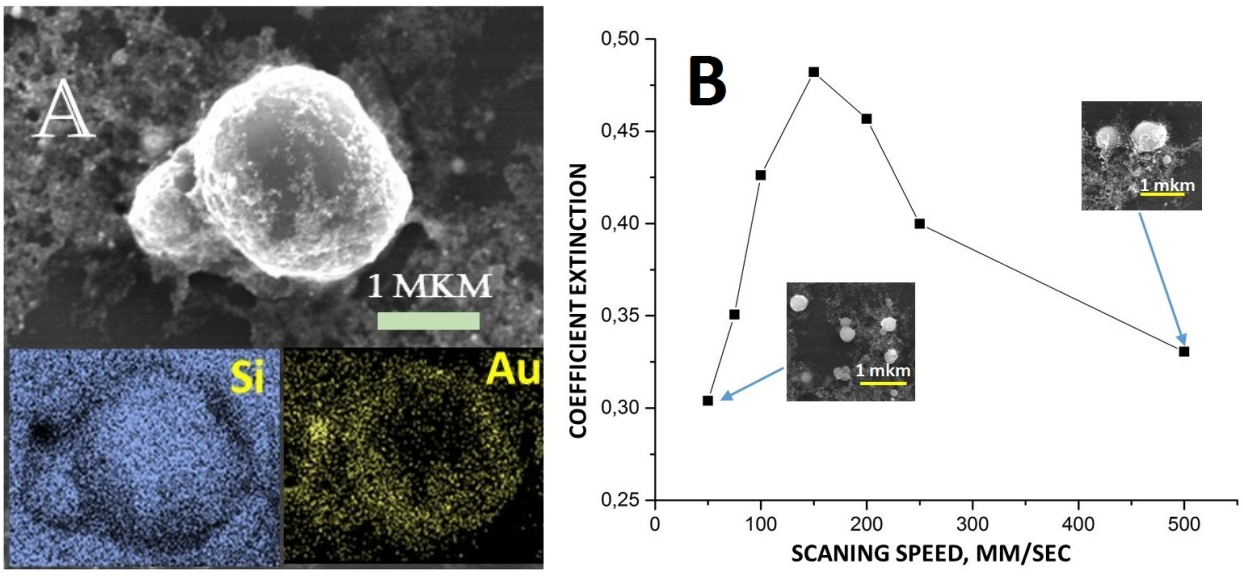

Figure 1: (a) SEM image hybrid Si-Au NP with elemental composition. (b) Extinction coefficient at wavelength 520 nm from laser skanning speed. At the inset SEM image hybrid Si-Au NP dried.

Laser nanoablation – a novel technique for presize structuring and functionalization of diamond

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The technique of low intensity multiple short pulsed laser ablation of diamond in air has been proposed and studied. It can be realized at pulse intensity below surface graphitization threshold with ablation rates in the range 10^{-7}–10^{-2}/pulse. Correspondingly, only few carbon atoms or clusters can be removed (in average) per each laser pulse. That is why such an ablation regime was called nanoablation.

Major features of nanoablation will be presented for femtosecond laser irradiation (wavelengths 800, 400 and 266 nm). For comparison data obtained with nanosecond pulses of excimer lasers will be given. Synthetic single crystals and polycrystalline diamond plates were used in the experiments. Interferometric control of laser induced diamond refractive index modification and material multi-photon ionization was performed.
The two-step nanoablation mechanism is discussed. In the first step, gradual (from pulse to pulse) lattice modification takes place as a result of laser-induced strong diamond ionization. Measured carrier concentration could reach 10^{21}–10^{22} cm^{-3}. In the second stage, at the sample surface weakly bonded carbon atoms are formed and etched away by thermal and photo oxidation stimulated by the sequence of laser pulses.

It will be demonstrated that nanoablation technique can be efficiently applied for presize diamond surface nanostructuring.

Another found important field of nanoablation application is formation of structural defects at the laser treated diamond surface. In nitrogen doped diamond crystals part of such defects was identified as nitrogen-vacancy (N-V) color centers. This is a new route to fabrication of single-photon N-V emitters in diamond that are of particular interest for quantum optics.

Inducing LIPSS by multi-pass and cross-directional scanning of femtosecond beam over surface of thin metal films

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During interaction of femtosecond laser beam with metal surfaces, laser induced periodic nanostructures, LIPSS can be formed, which may improve properties of materials. Having excellent mechanical properties, multilayer thin films, like 5x(Al/Ti)@Si, are convenient for forming of high quality LIPSS [1] due to their multilayer structure. We have exposed the multilayer thin film metal systems 5x(Al/Ti)@Si with femtosecond beam from the laser system Coherent Mira 900 in NIR with various scanning configurations [2]. The irradiated samples have been analyzed by Tescan Mira3 SEM. The beam scanned over the surface of the samples with multi-pass and cross-directional scanning configurations with the change of polarization direction. The formation of LIPSS is most probably due to the occurence of surface plasmon polariton, which leads to the periodic distribution of energy on the sample surface. The orientation of the LIPSS is related to the direction of the beam polarization. During multi-pass scanning, LIPSS maintained its configuration. The preservation of structures occured to some extent. Depending on the accumulated energy, two forms of LIPSS were generated: "hills", for less accumulation, and "trenches" for greater accumulation. "Hills" are non-ablative, probably are due to the build-up of the material and are parallel to the polarization direction. "Trenches" are formed by ablation and are perpendicular to the polarization direction. During cross-directional scanning, LIPSS of orthogonal directions have been generated. The value of the "hills" period was around 360 nm and the width was ∼285 nm. The values of "trenches" period fluctuated between 320 and 380 nm, while width was between 85 and 45 nm. Proposed mechanism is that, for less accumulated energy, "hills" formed, while more accumulated energy leads to the ablation and formation of "trenches".

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Registration of periodical structures in Ag-doped solgel films by interference of picosecond laser pulses

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Sol-gel films impregnated by metal salts or nanoparticles seems to be one of the possible prospective composite media for registration of optical patterns with high resolution [1]. The main attractive feature of the sol-gel films is the possibility of controlled introduction into their composition various impurities, including nanoparticles.

In this way, the aim of the work is a fabrication and investigation of registration mechanism and optical properties of laser-induced periodic structures formed into sol-gel films impregnated with a silver salt in view of interference pattern.

As the sample the thin porous SiO_2 film (h = 200 nm) with average pore size ∼7-10 nm deposited on glass substrate was used. Silver ions were introduced into the film by soaking in an aqueous solution of $AgNO_3$ (0.5M) for 40 minutes, and after it was dried at room temperature for 12 hours. Nd:YAG laser (355 nm wavelength with 25 ps pulse duration) was used as a laser source. Laser beam passed through the phase grating (with period p = 30 μm fabricated by laser-induced μ-plasma technology [3]) and divided into a two beams. The beams were overlapped after confocal scheme creating an interference pattern in the sample. Confocal optical system allowed to form the pattern with a period of ∼410 or ∼520 nm. Periodical structures were obtained at peak intensity 10^{10} W/cm^2 and the number of pulses 100—1000. The mechanism of registration such a pattern was investigated and seems to be combined from photochemical and thermophysical stages. Reflection and diffusion spectra show the correlation of peaks with the period of fabricated structures.

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Ultrafast laser nanofabrication of advanced nanophotonic structures

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In past decades, direct processing of different noble-metal materials using short- and ultrashort laser pulses has become a matured technology allowing rapid prototyping of various nanotextured morphologies. Currently, pulse repetition rate of the commercially available laser systems used in the nanotexturing experiments enters sub-GHz range ensuring extremely fast material processing at the speed reaching several cm^2 per second. From the other hand, laser nanofabrication with a "structured light", specially designed laser beams with a complex intensity profile allowing specific conditions of energy deposition into the irradiated materials becomes a hot topic in the area of laser nanofabrication. Here, we overview our recent results related to direct fabrication of various micro- and nanoscale structures and surface textures using short and ultrashort laser pulses. First, we present the results on formation of chiral surface nanoneedles under noble-metal films ablation with "perfect" vortex beams having variable topological charge as well as specially designed spiral beams discussing the underlying formation mechanisms and the ability to tune the structure's chirality. Then, we present the method allowing fabrication of various isolated plasmonic nanostructures with porous inner structure via the ablation of the nitrogen-doped noble-metal films. The proposed method allows to precisely tune the porosity of the produced structures through the control over the nitrogen content in the irradiated noble-metal films. Several applications of the presented laser-printed nanostructures and nanotextures in plasmon-mediated structural color generation, biosensing based on surface-enhanced spectroscopy effects, etc. are also discussed.

Manipulation by surface plasmon resonances: optical and materials aspects

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Nanopatterning of Si surfaces is a promising CMOS-compatible approach in fabricating Si nanostructures, which were recently demonstrated as a promising, easy-to-functionalize and quantum-confinement 2D-material platform for energy and hydrogen storage, planar nanoelectronics, thermoelectrics and optoelectronic applications. Meanwhile, novel efficient routes for large-scale fabrication of Si nanopatterns are currently required in lieu of the rapidly growing interest and developing research to harness their common and to explore novel functionalization modalities.

Advanced fabrication of ultrafine (sub-wavelength, $\Lambda \sim \lambda/3$, or deep-subwavelength, $\Lambda \sim \lambda/10$) surface nanoripples by ultrashort ($\sim$0.1-10 ps) laser pulses has proved for a long time to be a "green", fast technology for large-scale surface nanopatterning in the form of regular arrays of surface 1D-nanowires or 0D-nanodots, managing surface nanostructures by means of laser wavelength, polarization and fluence variations for tribological, nanophotonic and biomedical applications. In this way, ultrathin 2D graphene and diamond nanoripples emerged during ultrashort-pulse laser nanopatterning of highly oriented pyrolitic graphite or monocrystalline diamond, respectively. Mesoscale internal (sub-surface) crystalline structure of ultrafine surface ripples on different material surfaces (Si, InSb, Al) was thoroughly explored via their cross-sectional analyses by means of high-resolution transmission electron microscopy (TEM) in order to reveal their underlying material transfer mechanism – spallation, phase explosion, surface thermocapillary hydrodynamic melt flows, which are still controversary, comparing to the commonly accepted plasmonic origin of periodical interferential energy deposition on material surfaces. Moreover, so far for Si surfaces the only sub-micron broad and shallow ripples (1D-nanowires) were observed under laser nanopatterning in ambient air, comparing to ultrafine – down to sub-60 nm broad – surface nanoripples revealed during ultrashort-pulse laser nanopatterning of titanium surfaces in sub-ablative "dry" and "wet" ablative regimes, indicating a potentially promising low-fluence window for "wet" way 2D-nanopatterning of silicon. This is somewhat surprising since the effect of the ambient medium is commonly considered as the refractive index (n_a) scaling down for the exciting wavelength λ, $\Lambda \sim \lambda/n_a$, which is usually neglected for typical magnitudes $n_a < 2$ in optically-transparent materials. Moreover, other related secondary important issues of ultrafast energy transport and mass-transfer temporal scales emerge upon the primary prompt surface-plasmon mediated nanoscale fs-laser energy deposition, requiring novel fabrication mechanisms and experimental regimes to be explored in order to achieve ultimate spatial resolution during fs-laser patterning.

In our study, we report on comparative 1030-nm fs-laser experimental studies of multi-shot deep-subwavelength nanopatterning of Si surfaces in transparent liquid carbon disulfide and other solvents, in order to explore potential modalities of wet ultrashort-pulse

laser nanoscale processing in terms of resolution, robustness and throughput. The resulting quasi-regular surface plasmon-mediated Si nanopatterns (nanoripples) were comprehensively characterized by scanning electron microscopy (SEM), EDX, TEM, XPS, Fourier-transform (FT-IR) spectroscopy and Raman micro-spectroscopy and numerically FDTD-modeled in terms of quasi-static interferential laser-SP electrical field distributions, in order to unveil their nanoscale formation paths.

Control of periodic surface structures on silicon by combined temporal shaping and polarization control of femtosecond laser pulses

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The application of temporally shaped femtosecond laser pulses in the strong interaction of laser pulses with solid state surfaces has been a particularly interesting subject of research during the last decade. When strong and ultrashort laser pulses interact with a Silicon surface they excite a surface field called surface plasmon. This in turn interacts and interferes with the incident laser field and when the intensity of the interaction is strong enough, it results to creation of periodic micro- and nanostructures on the surface. The introduction of temporal pulse shaping of the incident laser pulses has allowed the fine tuning of the morphological characteristics of the generated micro- and nano-structures. This has been ascribed to the control, to a certain degree, of the fundamental mechanisms which follow the laser stimulus. This control has been achieved through the manipulation of the width and generally the shape of the temporal profile of the incident laser pulses.

Further on, in this presentation we will discuss our extension to our past work on the subject where we are combining control of the temporal profile combined with control of the polarization states of the temporally shaped laser pulses. By this we demonstrate the capability to exercise advanced control on the laser-induced periodic surface structures (LIPSS) on Silicon by combining the effect of temporal shaping, via tuning the interpulse temporal delay between double femtosecond laser pulses, along with the independent manipulation of the polarization state of each of the individual pulses. For this, cross-polarized as well as counter-rotating double circularly polarized pulses have been utilized. The pulse duration was 40 fs and the central wavelength of 790 nm. The linearly polarized double pulses are generated by a modified Michelson interferometer allowing the temporal delay between the pulses to vary from -80 ps to +80 ps with an accuracy of 0.2 fs which allows fine tuning of the optical delay between the two laser electromagnetic fields. We show the significance of fluence balance between the two pulse components and its interplay with the interpulse delay and with the order of arrival of the individually polarized pulse components of the double pulse sequence on the final surface morphology. For the case of counter rotating circularly polarized pulses we found that when the pulses are well separated in time then the surface morphology attains no axial symmetry. But strikingly, for the case of the cross polarized pulses when they both temporally overlap, we demonstrate, for the first time in our knowledge, the detrimental effect that the phase delay has on the ripple orientation: it exhibits a 90 degree rotation for every half-cycle of optical field delay between the two laser pulse components. Our results provide new insight showing that temporal pulse shaping in combination with polarization control gives a powerful tool for drastically controlling the surface nanostructure morphology.

Nanoscale spatial and femtoscale temporal characterization of laser pulses

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We report results of our investigations demonstrating a nanoscale, ultrafast, and multiorder optical autocorrelator with a single plasmonic nanostructure for measuring the spatio-temporal dynamics of femtosecond laser light. As a nanostructure, we use a split hole resonator (SHR), which was made in an aluminum nanofilm. The Al material yields the fastest response time (100 as). The SHR nanostructure ensures a high nonlinear optical efficiency of the interaction with laser radiation, which leads to (1) the second, (2) the third harmonics generation and (3) the multiphoton luminescence, which, in turn, are used to perform multi-order autocorrelation measurements. The nano-sized SHR make it possible to conduct autocorrelation measurements (i) with a subwavelength spatial resolution and (ii) with no significant influence on the duration of the laser pulse. The time response realized by the SHR nanostructure is on the time scale of about 10 fs. In the UV wavelength range, the time response of the nanoprobe may increase to 100 as.

Controlling of plasmon damping on nonmetallic gratings excited with intense femtosecond laser pulses

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In 1902, Wood discovered the appearance of unexpected bright and dark lines, the so-called *Wood's anomalies*, in the reflection spectrum for a metallic diffraction grating illuminated by a continuum light source [1]. Nowadays, it is well known that the phenomenon is attributed to surface plasmon polaritons (SPPs) excited at the interface between a periodically grooved metal surface and a dielectric material (air). As the excitation of SPPs is a unique technique to localize photon energy on the nanoscale, increasing attention has been focused on potential applications such as gas and biosensors, waveguides, extraordinary optical transmissions, high-resolution imaging, light-emitting diodes, photovoltaics, and photodiodes.

Recently, the excitation of SPPs has also been proposed as the mechanism responsible for the formation of laser-induced periodic surface structures (LIPSSs) that were observed for a variety of solid surfaces under irradiation of intense femtosecond (fs) laser pulses [2]. It is considered that SPPs would play a dominant role in LIPSS formation even on nonmetallic materials such as insulators and semiconductors, as the intense fs laser pulse ionizes the target material to form a thin metal-like layer on the surface. While the excitation of SPPs on solid surfaces is expected to provide a promising approach to laser precision processing of materials, the mechanism based on SPPs is still being debated. The ambiguous points on the role of SPPs in LIPSS formation arise from the fact that the proposed mechanism has mostly been based on the ablation traces left at the target surface, and experimental studies reporting definite or direct evidence for the excitation of SPPs, in particular, on a nonmetallic target irradiated with fs laser pulses, are lacking.

Using a reflectivity measurement, we have reported the experimental observation of surface plasmon resonance (SPR) on a nonmetallic material with a grating structure excited by intense *p*-polarized 100-fs laser pulses [3]. Figure 1 shows a schematic drawing of the optical configuration to measure the reflectivity R at the Si grating surface. For the pump light we used linearly polarized, 100-fs laser pulses at a wavelength $\lambda \sim 800$ nm from a Ti:sapphire laser amplifier system. The fs pulse at the fluence F = 500–2000 mJ/cm^2 was irradiated to the Si grating surface. The results are shown in fig. 2 [4]. R measured with the *p*-polarized fs pulses exhibits an abrupt decrease to create a sharp minimum at $\theta = 23.0$–$24.0°$, or SPR curves. The width $\Delta\theta$ of the SPR curves is plotted in Fig. 3 as a function of F, where $\Delta\theta$ is defined by the full width at half maximum (FWHM) of the curve measured. The result shows that $\Delta\theta$ decreases monotonously from $\sim 4°$ to $\sim 2.5°$ in an increase of F from 500 mJ/cm^2 to 2000 mJ/cm^2.

The results show a reduction of the plasmon damping on Si gratings in increasing the laser fluence of the fs pulses. The experimental and theoretical results also provide direct evidence that the internal damping is the dominant decay process of the surface plasmon polaritons as compared to the radiation damping, and decreases with the fs laser pluses at a higher fluence, owing to an increase of the conductivity of the surface with the higher density of the electrons produced. These findings show that the plasmon damping on Si gratings can be controlled with the laser fluence of the fs laser pulses.

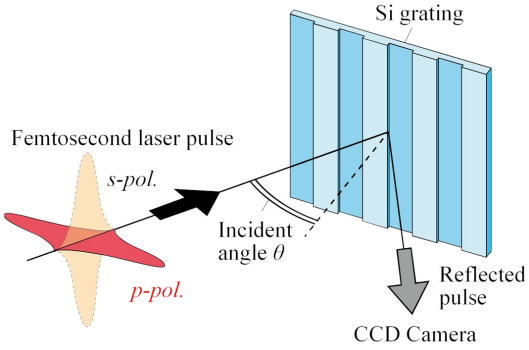

Figure 1: Schematic diagram of the optical configuration for the reflectivity measurement.

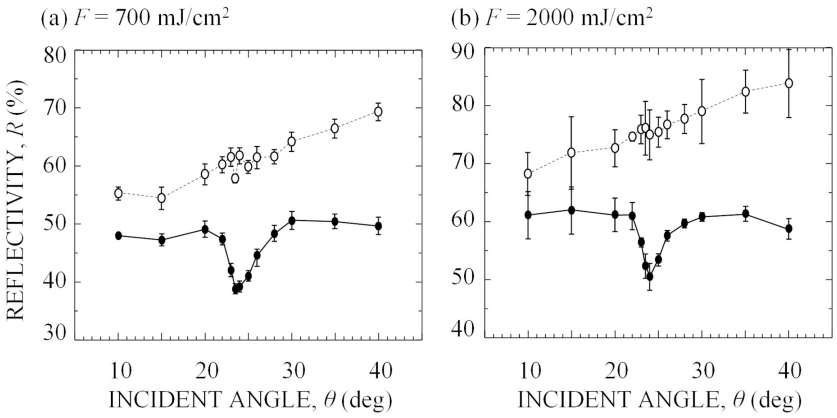

Figure 2: Reflectivity R for the Si grating measured as a function of the incident angle θ of the fs laser pulse at (a) F = 700 mJ/cm^2, and (b) F = 2000 mJ/cm^2, where the solid and open circles correspond to R observed with p and s polarization. The solid and dashed lines simply connect the data points for easier tracking of the dependence.

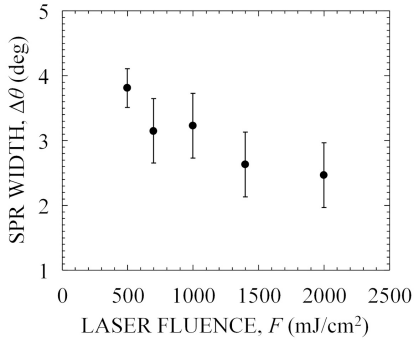

Figure 3: SPR width $\Delta\theta$ measured from the reflectivity for the Si grating observed as a function of the incident angle θ of the fs laser pulse at F = 500–2000 mJ/cm^2.

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Writing of crystalline tracks in glass by Laguerre-Gaussian femtosecond laser beam

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The method of local crystallization of glasses by a femtosecond laser beam has been actively developed in recent years and has recently allowed the writing of single-crystal channel waveguides in glass [1, 2]. One of the problems of development of such waveguides is the complex structure of the laser-written crystalline track, which is generally formed by two growth fronts and has a horseshoe-shaped cross-section with an amorphous or polycrystalline interlayer between the two crystalline parts. This feature of crystal growth determines the multimode composition of the crystalline channel waveguide laser-written in the glass and makes it difficult to develop single-mode single-crystal waveguides.

In this work, experiments on the space-selective crystallization of glass by a Laguerre-Gaussian femtosecond laser beam (LG_{01} mode) obtained using a liquid crystal spatial light modulator HAMAMATSU LCOS-SLM were carried out for the first time., a femtosecond laser TETA-100 (Avesta Ltd.) generating pulses with energy up to 3 μJ at a wavelength of 1030 nm was used to write crystalline tracks inside lanthanum borogermanate glass. The pulse repetition rate and duration were 100 kHz and 300 fs, respectively. Laser beam was focused in the glass bulk at the depth of 0.18 mm by means of Olympus LCPLNIR objective lens (N.A. = 0.45). Extended crystalline tracks consisting of $LaBGeO_5$ ferroelectric phase were grown from a seed crystal preformed by a stationary laser beam. The minimal pulse energy providing the continuous growth of the crystalline track is found to be higher in the case of the Laguerre-Gaussian mode as compared with the conventional Gaussian beam (2.25 μJ vs. 1.80 μJ).

A comparative analysis of the morphology of crystalline tracks grown in glass by the Gaussian laser beam and a Laguerre-Gaussian beam has been performed. The Laguerre-Gaussian beam is shown to enable the formation of a crystalline track with a homogeneous cross-section that is not divided into two parts by a polycrystalline interlayer, which is presumably associated with a more uniform temperature distribution in the glass bulk provided by the LG_{01} mode in contrast to Gaussian one. This result opens a possibility to improve the characteristics of the crystalline channel waveguide in glass.

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Laser-induced coloration of metals surface

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Surface laser modification, the creation of micro- or nanoscale topologies on the metals surface, is important for various fields of science and industry, such as laser marking (including color marking) [1], biomedical applications [2] and tribological characteristics monitoring [3]. In addition, product labeling is important not only for identification purposes, but also for authentication, that is, as an advanced technology of protection metal products against falsification.

The technology of metal surfaces coloring, based on laser-induced surface structuring, is shown. The visible light interaction with a structured surface is used to optimize the light reflection and to create a structural color. It is known that a number of structural colors can be created with the help of the following mechanisms: interference, diffraction, scattering, surface plasmon resonance [4]. In this paper, the metal surface coloration effect is demonstrated by the spatial periodic structures (LIPSS) formation with a period of the order of radiating wavelength 1 μm. Due to dispersion on the grating, white light is decomposed into a spectrum. Depending on the grating geometry, this leads to characteristic structural high brightness colors. The structures orientation controlling is achieved due to the laser radiation polarization direction controlling. Due to what is possible to produce the color elements, smoothly changing color gamut, and various dynamic effects at different viewing angle. The second demonstrated in this work staining mechanism is related to the surface plasmon resonance effect on metal randomized particles. The work shows the method of silver staining due to the formation of silver nanoparticles (NP) on its surface. As a result of laser irradiation of silver, particles of 10 to 50 nm in size form on its surface, depending on the regimes of laser radiation action, the average size and surface density of NPs vary. Depending on the exposure parameters, different colors are observed on the surface. Also, the effect of metal blackening is observed, due to numerous absorption during iterative reflection.

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Ultrafast dynamics of magneto-optical effects in nanostructured media with artificial dispersion

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The active control of optical signals in the time domain is a main driving force for the fast all-optical information processing nowadays. Nanostructured media modify the group velocity and slow the light down, as the artificial light dispersion emerges. Resonant changes in magneto-optical response of these systems is the result of the increased light-matter interaction time. Non-trivial time-resolved dynamics of the effects can be observed on the femtosecond timescale, which is determined by the lifetime of the excitations. In this work we show that layered materials, like magnetophotonic crystals, allows one to realize modification of the polarization state inside a single femtosecond laser pulse (see fig.1(a)). For example, Faraday rotation of the leading part of the pulse can differ from that of its tail and from a steady-state value. Moreover, Faraday rotation can both increase and decrease in time depending on interference conditions for a particular pulse central wavelength. The use of magnetoplasmonic crystals opens a new way for magnetic-field-controlled pulse shaping of a single femtosecond laser pulse (see fig.1(b)). The shape of the pulse, reflected from a magnetoplasmonic crystal, is different for opposite directions of an external magnetic field. The reason for that is modification of a surface plasmon-polariton dispersion law, when external magnetic field is switched on.

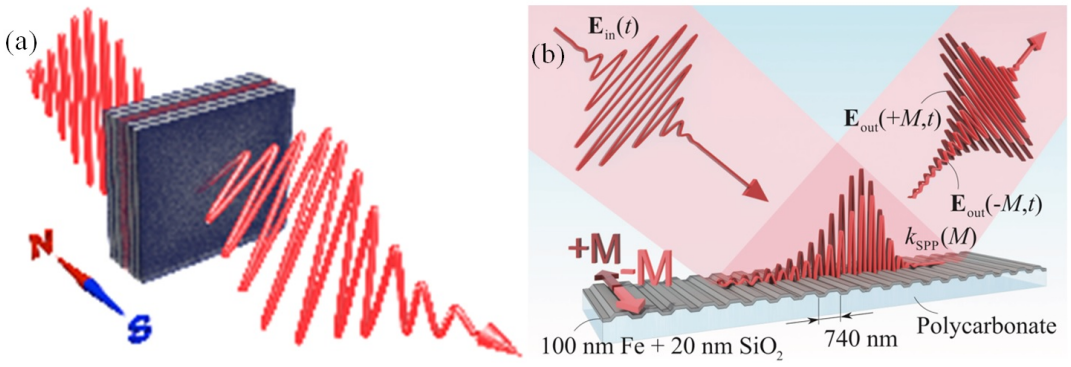

Figure 1: Femtosecond intrapulse evolution of polarization plane rotation due to Faraday effect in magnetophonic crystals. (b) Ultrafast pulse shaping by magnetoplasmonic crystals [1,2].

The work is supported by Russian Foundation for Basic Research (grants № 17-52-560011, № 18-52-50021).

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Nanostructures in lattice fabricated by interference laser processing technique

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Area processing technique is the key for the fabrication of functional surfaces and devices. We have developed interference pattern processing technique for the fabrication of nano-sized 3D structures in lattice such as nano-whisker [1,2], nano-drop [3], designed pattern formation [4,5], as shown in Figure 1. These structures has a variety of applications such as plasmonic devices, metamaterials etc.. This method can fabricate a variety of nanostructures instantaneously without post process in atmospheric condition. So, comparing to figuring methods such as FIB and lithography, this can be better alternative. In this paper, we will summarize the background, past and recent results of nanofabrication. In addition, control of interference pattern and beam shaping for uniform area processing will be shown.

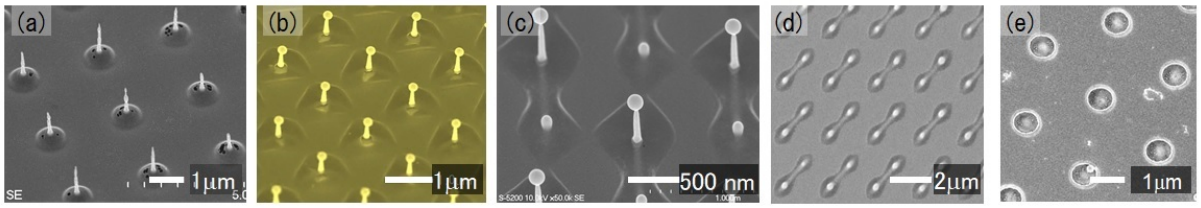

Figure 1: Gold nanostructures in lattice fabricated by an interference pattern of a single shot of femtosecond laser. (a) nano-whisker in square lattice, (b) nano-drop, (c) duplex structure of nanodrop and nanobit, (d) designed structure (bit pair), and (e) hole in triangle lattice.

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Preparation of bimetallic nanoparticles by laser ablation

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At the present time, much attention is paid to the study and synthesis of composite metallic nanoparticles due to their unique physical, optical and catalytic properties, which differ significantly from characteristic for bulk materials [1-2].

In this paper, bimetallic nanoparticles were obtained from thin films by laser ablation in water and by the dewetting. The films were deposited using a magnetron sputtering installation in an argon atmosphere. Two-layer films of metals were used as targets. As the metals were chosen: Ag, Au, Al.

The source of radiation was a fiber laser marker HTF MARK (Bulat) on Yb^{3+} ions with pulse duration at half-height of 120 ns, maximum pulse energy of 1 mJ and pulse repetition rate up to 80 kHz. The radiation was focused by a galvanoscanner with a lens focal length of 160 mm. The optical properties of the obtained colloidal solutions were studied, and the nanoparticles were visualized by scanning electron microscopy (SEM). The chemical composition of the obtained bimetallic nanoparticles was confirmed by energy-dispersive x-ray spectroscopic chemical microanalysis (EDS) (Fig.1).

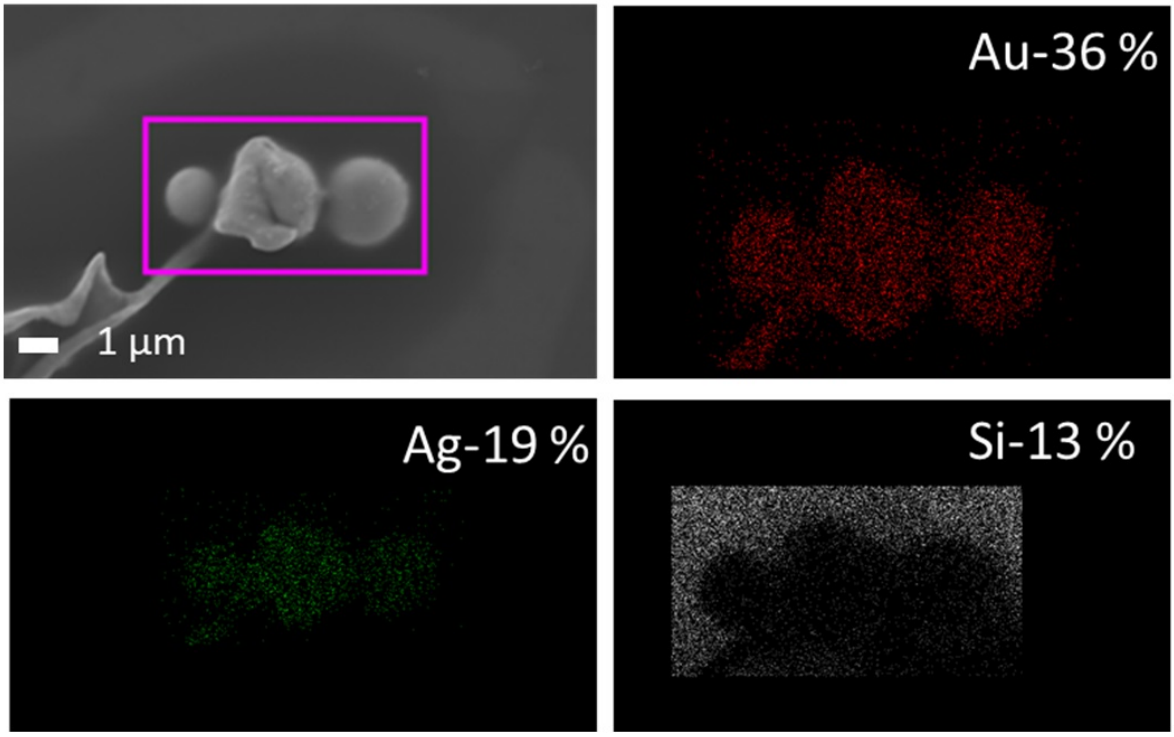

Figure 1: EDS analysis Ag@Au nanoparticles

This work was financially supported by the grant of the Russian Science Foundation (project № 18-15-00220).

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Structured Materials by Ultrafast Vortex Pulses Illumination

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Optical vortices [1], i.e. helical light beams, exhibit unique physical properties, e.g. helical wavefront (i.e. handedness), annular intensity profile, and orbital angular momentum (OAM), therefore, they exploit many new fundamental light-matter interactions. In fact, optical vortices have been attracting in various fields, such as optical tweezers/manipulations, optical space/fiber telecommunications, quantum communications/informations, and high spatial resolution fluorescence microscopes. In recent years, we proposed laser materials processing based on pulsed or continuous-wave (CW) optical vortex illumination, in which optical vortices twist the irradiated materials to establish chiral structured materials on a nano-/micro-scale owing to OAM transfer effects [2,3]. To date, various structured materials, such as chiral metal needles, chiral polymeric surface reliefs, and twisted photo-polymerized fibers, have been successfully fabricated by employing optical vortex illumination.

In particular, ultrafast vortex pulses force strongly the irradiated materials to direct towards a dark core of the optical vortices, so as to form unique structured materials, for instance, monocrystalline metal/silicon needles or spheres [4,5]. The ultrafast vortex pulses also enable us to induce local mass transport of polymers and polymerization with the help of multiphoton absorption, thereby resulting in the establishment of sub-wavelength scale chiral surface surfaces (with a size beyond the diffraction limit) and millimeter scale helical polymer fibers. Furthermore, ultrafast optical vortex pulses will structure spiral ripples, so-called spiral laser-induced periodic surface structures on the materials [6].

Such ultrafast optical vortex materials processing will potentially pave the pathway towards novel advanced technologies of material sciences: for instance, development of the chiral plasmonic devices, the ultra-highly sensitive detector and collector for the chiral chemical composites at high time and cost efficiencies, and short-distance optical waveguides for optical vortex modes.

In this presentation, we review the state-of-art of the chiral nano-/micro-structures formed by the illumination of ultrafast optical vortex pulses. We also review recent progress of advanced ultrafast optical vortex sources for materials processing.

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Use of femtosecond laser exposure to create topological structures in high-temperature superconducting composites

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In this paper presents the results of a study the possibility of creating topological structures in second generation (2G) HTS tapes. The widespread application of second-generation HTS tapes due to the possibility to use liquid nitrogen cooling. The material have high current-carrying characteristics and strength properties. However, an important problem is still remains: we need to increase the critical current, reduce the losses in the superconductor with alternating current, and also reduce the time for the transition of the superconductor to the normal state under the action of the current pulses. The latter is particularly important for superconducting current limiters. One of the approaches to improving the characteristics of superconductors is the separation of the film into thin filaments. Modern superconducting film superconductors (coated conductors) are composite materials consisting of a Ni-W metal substrate, buffer layers, a HTS film layer and protective coatings of copper and silver. The thickness of the superconductor film is 1 μm.

To create a topological structures we used laser system FEMTO-T, developed in NRNU MEPhI. The pulse duration was 2 ps, the pulse energy varied from 100 to 850 nJ. To create a spatial pattern on samples complex is equipped with a focusing and scanning system with a minimum step is 150 nm. The application of ultrashort laser pulses allows to remove superconductor material without heating and degradation of the surrounding area. Due to this feature the minimum width of cut does not exceed 1 micron.

With the help of a laser complex a modification of industrial high-temperature superconducting tapes of the second generation was performed. Two types of topological structures were created: a triangular lattice of through holes and longitudinal sections of the film forming individual filaments. Structural studies of individual defects and cuts were carried out. Dependences of the depth of the holes and cuts and the diameter of the hole (the width of the cut) on the energy of the pulse and the number of pulses per point are obtained. It were found the laser exposure regimes for the complete superconductor film separation into individual filaments. The current-carrying characteristics of the original and modified HTS tapes, the dependence of the critical current on the magnetic field, and the magnetization curves were studied. Investigations of the effect of created topological defects on transient processes under the influence of current pulses were also carried out. The positive effect of the modifications of HTS tapes is shown.

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Phase-change magnetic memory by ultrafast laser patterning of Fe-Al alloys

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It is well established [1,2] that laser pulses are able to reversibly transform so-called phase-change alloys between their states with different structural order, thereby altering their electrical resistivity and/or optical reflectivity. We demonstrate [3] that short-pulsed lasers induce a reversible transition in the thin-film (40 nm) Fe60Al40 alloyed system from its non(ferro)magnetic to ferromagnetic state. We argue that the observed phenomenon is explainable in terms of the order (B2)–disorder (A2) chemical transformation. A high-intensity pulse causes melting at 1700 K of the initially nonmagnetic, structurally ordered, material and its disordering. Upon cooling, the disorder persists in the undercooled melt down to a low enough temperature (1100 K) below which the resolidified alloy is not able to reorder and appears to be ferromagnetic after cooling down. However, a subsequent low-intensity (below melting) pulse restores the order and nonmagnetic state. Such cycling of the ferromagnetism by laser can be considered as a novel approach to high-density magnetic recording [4,5]. The results obtained in this study pave a route to solving the problem of thermal fluctuations of magnetization, which give a fundamental limitation to storage data density in conventional magnetic recording.

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Design of diffractive optical elements for laser fabrication of U-shaped element arrays

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We propose a new modification of the well-known iterative Gerchberg-Saxton algorithm that can be used to calculate a pure-phase transmission function of diffractive optical elements which generate complex intensity distributions with submicron features. The proposed algorithm allows us to design an element shaping intensity distributions that can be used to create a nano/micro U-shaped element with a single femtosecond laser pulse. The size of the fabricated elements depends on the numerical aperture of the utilized microobjectives and can be less than one micron. We demonstrate results of laser fabrication of such U-shaped element arrays on the surface of glass substrates with different deposited metals such as gold, silver, and aluminium. We also provide microscopy and spectroscopy analysis of the manufactured U-shaped elements.

Tighter focus for ultrashort pulse vector light beams

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We perform a comprehensive study of focusing of ultrashort/single-cycle optical beams with linear, circular, radial and azimuthal polarizations with and without a vortex while comparing them with focusing of long pulse beams. We show that in all the considered cases the focal spot of short pulses decreases significantly, in accordance with previous studies of ultrashort pulse scalar beams and radially polarized ones, supporting the conclusion that the focal spot decrease is caused by increased bandwidth with higher frequencies dominating the spectrum. More significantly, we show that, upon pulse shortening, some input polarizations exhibit larger degree of focus tightening when compared to other polarizations. We demonstrate that this is due to stronger dependence on/heavier weighting by frequency of those focused vector beam components which are proportional to the 1st order Bessel function, J_1, as compared to the components proportional to zero order Bessel function, J_0. This finding opens new paths to control the focusing of ultrashort, large frequency bandwidth beams by emphasizing the frequency-dependent contribution of certain order Bessel components vs other ones using, e.g. phase apodization of the incoming beam.

Nanoparticles fabricated by pulsed laser ablation in liquids and their applications in biomedicine

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Pulsed laser ablation in liquids is an emerging field, particularly suitable for the generation of nanoparticle colloid without utilization of organic stabilizer ligands and with an outstanding material variety [1]. In this work the basic principles of this technique will be shortly reviewed, including cavitation bubble dynamics and to what extent they may influence NP formation [2]. This is followed by a detailed discussion on the chemistry of the resulting particles. Particular focus will be on the interaction of PLAL-fabricated NP with inorganic ions and their impact on colloidal stability and particle size control [3-5]. Another main point will be on bimetallic nanoparticle, in particular alloys with high miscibility (e.g. AgAu) [6,7] as well as those with partial miscibility which form segregated structures (e.g. FeAu) [8]. In this context, we will discuss the correlation between the internal phase structure of the resulting particles and the composition of the target and the solvent. Finally, some examples as to the application of laser-generated nanoparticles in biomedicine will be given, addressing the fields of nano-bio-conjugates [9,10] and nanocomposites [11,12].

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Ultrashort laser ablation of dielectrics

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Ultrashort femtosecond pulses provide highly-controlled interaction with dielectric materials. For single-shot ablation, intensities exceeding 10^{14} W/cm^2 at the target surface are easily reached with a focused microjoule pulse, yielding important benefits for ablation: low fluence threshold, strong determinism, high ablation efficiency and small residual affected zone. Decreasing the pulse duration down to few-cycle laser pulses, i.e. in the $\sim$10 fs range, is likely to further enhance the capabilities of direct femtosecond laser machining in the context of micro-nano-fabrication.

However, such intensities often require vacuum environment for proper propagation and focusing, because working in air ambience may rapidly yield significant nonlinear spatial distortion of the beam in the pre-focal region, or even air ionization, which inevitably hampers the applicability of such ultrashort pulse. We first analyze the beam characteristics and discuss the inherent limitations to ablation of the surface of dielectrics [1]. Then we show the existence of an energy regime, above the ablation threshold, where the beam is still unaffected, authorizing proper ablation in air. Moreover, even in a high-intensity regime (i.e. with a strongly self-reshaped beam) we demonstrate that prediction and realization of high-quality ablation craters are still attainable in air, provided that the fluence is properly calibrated [2].

Besides, we develop complementary experimental diagnostics (beam-depletion and front-surface reflection measurements) that provide information on the created plasma and its transient dynamics. The global picture is that a 10-fs pulse duration yields high efficiency of absorption (several tens of percent at the threshold), while maintaining a low level of self-induced reflectivity. We compare and discuss the results acquired on fused silica SiO_2 and sapphire Al_2O_3, two dielectrics which have similar bandgaps but different absorption behaviors (in particular regarding electronic avalanche process) and we link the short-timescale plasma characteristics retrieved from our experiments to post-mortem morphological features of ablation craters.

This work therefore provides fundamental and practical information in the few-cycle-pulse ablation regime, exploring the transfer of laser energy to the material in connection with final ablation outcomes.

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Laser ablation thresholds of metals and semiconductors in air and liquid media during fs/ps micromachining

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The effect of pusewidth is known to have a great impact on the underlying mechanisms of laser ablation, as it may strongly correlate with the electron-phonon coupling of the material, thus influencing the ablation fluence threshold values. In our work we report the study of laser pulsewidth effect on the ablation thresholds of bulk gold, silver, silicon and selenium in air and liquid media (deionized water, isopropyl alcohol). Yb^+-doped ultrashort fiber laser system Satsuma, Amplitude Systhemes (λ =1030 nm, average energy up to 6 μJ) with gradually tuned laser pulsewidth τ from 0.3 to 10 ps was used to acquire single-shot microcraters on the surface of bulk materials, which were then measured via scanning electron microscopy. The resulting laser power varied from 0.6 to 20 MW and therefore the liquid layer of 2 mm was found to induce multifilamentation, resulting in the characteristic well-shaped submicrocraters (e. g., typical critical self-focusing power in water P_C(800 nm)≈2 MW [1]).

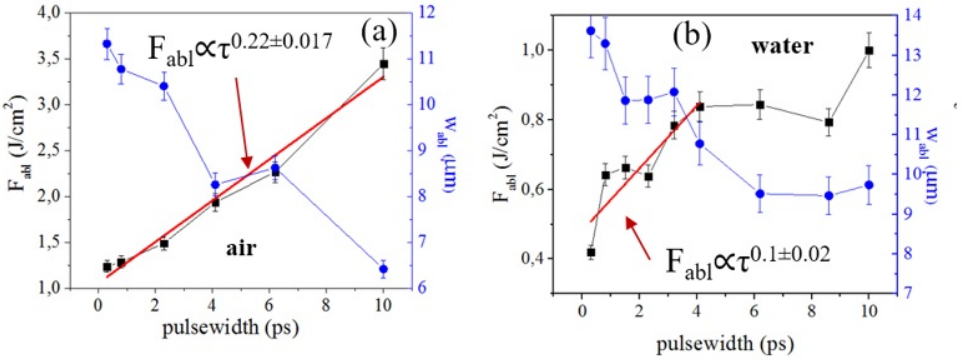

Figure 1: Threshold values F_{abl} and 1/?-radii w_{abl} for single-shot ablation of (a) bulk Au surface in air and (b) Ag surface in deionized water versus laser pulsewidth τ.

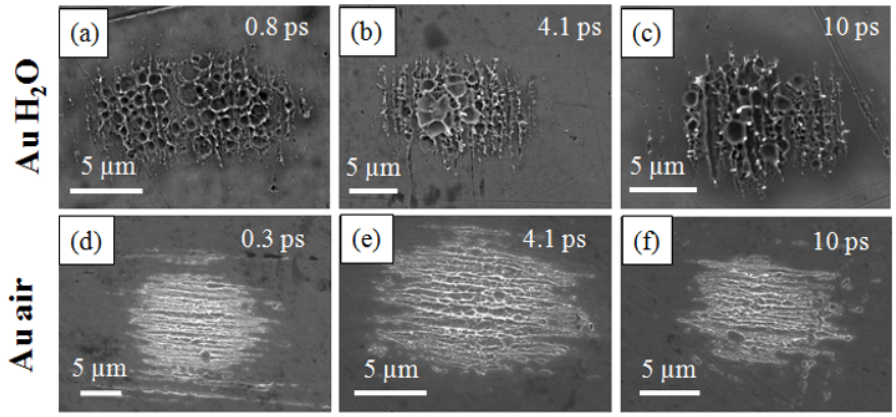

Figure 2: Single-shot ablation craters on the surface of bulk Au in water (a, b, c) and air (d, e, f), acquired at different laser pulsewidths (marked on the images).

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Formation of silicon nanoparticles by pulsed laser ablation of porous silicon in liquids

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Silicon (Si) nanostructures attract scientific interest for their possible application in the biomedicine because of the properties of Si itself (biocompatibility, biodegradability and low toxicity), and also due to the possibility of changing the semiconductor properties when switching to nanoscale. For example, small-size nanoparticles (less than 7 nm) demonstrate quantum-size effects that lead to effective photoluminescence (PL) in the visible and near infrared range, corresponding to transparent diagnostic window of the biotissue (700-1300 nm), which makes it possible to use such nanoparticles as photoluminescent markers inside the living organisms for medical diagnosis [1]. For such applications it is important to develop fabrication methods, which allow obtaining Si nanoparticles with small size, desired optical properties and minimal chemical contamination simultaneously.

To solve this task, in this work, Si nanoparticles are formed by a hybrid approach that includes sequential application of the electrochemical etching of Si in a hydrofluoric acid solution ($HF:H_2O:C_2H_5OH$ = 2:3:5) and pulsed laser ablation of etched Si wafers in various liquids (distilled water, liquid nitrogen, ethanol). Femtosecond (1250 nm, 180 fs, 10 Hz, 10 J/cm^2) and picosecond (1064 nm, 30 ps, 10 Hz, 17 J/cm^2) laser pulses were used for ablation. As a result of etching, porous Si is formed, consisting of Si nanocrystals and air pores. Such structure demonstrates effective photoluminescence. As a result of laser ablation of the porous silicon, nanoparticles suspensions with narrow distribution in size are formed.

Atomic force microscopy and scanning electron microscopy revealed the large fraction of nanoparticles with size less than 20 nm, which shows higher efficiency of the proposed two-stage method compared to single-stage laser ablation or mechanical grinding. Raman spectroscopy revealed red shift and broadening of the crystalline Si TO phonon mode, indicating the presence of a quantum-size effect in the formed nanoparticles. PL spectra of the nanoparticles produced in liquid nitrogen and ethanol demonstrate maximum at 746 and 720 nm correspondingly, which is within biotissue transparency window. On the other hand, nanoparticles produced in distilled water showed absence of PL, which is probably explained by formation of oxygen and hydrogen defects in the nanoparticles during ablation.

It was also demonstrated that PL lifetime of the Si nanoparticles ensembles produced in liquid nitrogen and ethanol increases by more than an order of magnitude after laser ablation. Obtained characteristics of the fabricated Si nanoparticles suspensions allow us to conclude that such objects can find application as photoluminescent markers for biotissue objects.

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Periodic surface structuring of fused silica and ULE glass using femtosecond laser pulses

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Femtosecond (fs) lasers have proven to be an excellent tool for processing of glass materials, as they are able to produce high quality ablation craters [1, 2] and volumetric modification [3] due to a reduced heat affected zone as compared to ps and ns pulsed lasers. Besides micromachining, drilling and volumetric modification, fs-lasers have also appeared to be efficiently applicable in fabrication of various kind of laser induced periodic surface structure (LIPSS) [4]. For different scientific applications it is desired to have LIPSS produced on large surface area with controlled periodicity. In this work, we will present results of a systematic study of the LIPSS produced on fused silica (FS) and Ultra-Low Expansion (ULE) glasses having different thermophysical and optical properties. The irradiation was performed using IR fs-laser in the fluence range close and well above the ablation thresholds of these two materials. We have systematically investigated the morphology of LIPSS formed in single- and multi-pulse regimes for both fixed irradiation spots and upon scanning the sample in raster fashion under linearly polarized focused laser beam.

In the case of ULE glass, microscopy results show the formation of coarse LIPSS with their periodicity close to the laser wavelength and orientation perpendicular to the polarization of the incident laser beam. For high-purity FS at the same irradiation conditions, a more complicated pattern of LIPSS is produced where local regions with both perpendicular and parallel orientation and even the coexistence of the two orientations are observed. The physical mechanisms responsible for this difference will be discussed.

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Femtosecond laser direct writing of depressed cladding waveguide in tellurite glass

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Good viscoelastic characteristics, mechanical strength, wide transparency spectrum and high nonlinear refractive index make tellurite glasses extremely attractive materials for fabrication of waveguiding optical devices, operating in IR and mid-IR. They are characterized by low phonon energy (with respect to silica glasses), and can be obtained with low 3d-transition metals and hydroxyl groups impurities content, which provides high transparency in the near and mid IR [1,2]. We performed a femtosecond laser direct writing of a waveguide in the bulk of tungstate-tellurite oxide glass doped with bismuth oxide. Generation of a supercontinuum in the mid-IR spectral region was investigated in the inscribed waveguide.

The depressed cladding channel waveguide was inscribed in the glass sample using the femtosecond laser operating at 1030 nm. Laser beam with pulse duration of 180 fs and repetition rate of 180 kHz was focused by microscopic objective with numerical aperture NA = 0.85 under the surface of glass sample, which was translated with velocity of 6 mm/s perpendicular to laser beam propagation. Under pulse energy of 70 nJ we recorded a set of homogeneous tracks forming the cladding with decreased refractive index. The cladding consisted of 32 tracks surrounding the unmodified core forming a waveguide with length of 14 mm. The single mode propagation of CW with loss of 0.15 dB/cm at 1064 nm was observed in linear regime.

Spectrum of femtosecond pulses generated by OPA laser system in the range 1000–2000 nm as it passed through the waveguide was studied. A laser beam was coupled to the waveguide from the polished end using a two-lens system. The laser light transmitted through the waveguide was collected from the exit end of the sample and analyzed using FTIR spectrometer. Non-linear propagation of the femtosecond pulses results in strong spectrum broadening. The phenomenon is explained by self-phase modulation triggered by large non-linear refractive index of the glass under study [3] and by Raman shift.

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One-pulse micro-ablation of steel by ultrashort laser pulses of varying duration

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To date, it is known that laser systems with nanosecond and more duration significantly lose in quality and efficiency of surface treatment of materials, in comparison with lasers with ultrashort duration (USLP) in the pico-femtosecond range [1]. At the same time, there are contradictory information for the USLP about what pulse durations in the pico-subpicosecond range are better for processing a given material. One of the key parameters in the laser processing of materials is the threshold value of the energy density F_{th}, the change which essentially depends on the pulse duration [2,3].

In the work, one-pulse processing of fresh areas of the steel polished surface of was carried out. The surface was processed by the second harmonic (515 nm) of the Satsuma fiber laser (Amplitude Systems). The pulse duration was varied via an output compressor in the interval 0.3–12.3 ps. Laser radiation was focused on the surface through a lens with a numerical aperture NA = 0.65. The morphology of the craters was visualized using a scanning electron microscope and an optical profilometer. The nonmonotonic dependence of the ablation thresholds and the characteristic radius 1/e of the ablation region w_{abl} is explained by the achievement of the electron-phonon thermalization time of the absorbed energy for a duration of ≈1.5 ps (Fig.1).

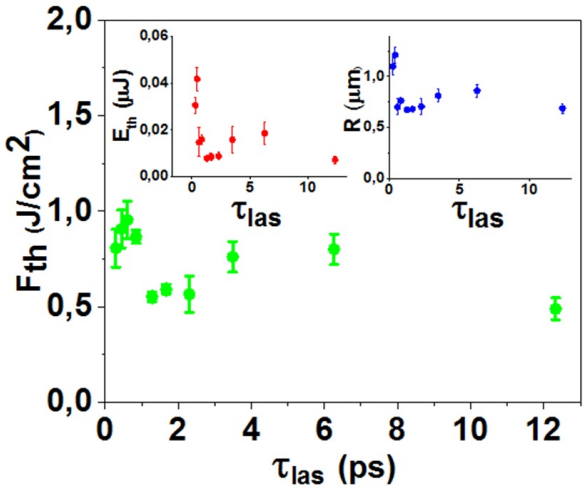

Figure 1: The threshold energy density F_{th} on the pulse duration, Insets: top left is the 1/e ablation radius W_{abl}, the top right is the threshold energy E_{th} in the pulse on the duration.

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Active and Nonlinear Semiconductor Metasurfaces

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Optical metasurfaces composed of designed semiconductor nanoresonators can provide exquisite control over polarization, spectrum and wavefront of light fields [1]. Usually, in order to generate light fields with desired properties, metasurfaces rely on their illumination by a known incident field, which is transformed into the desired light field upon transmission or reflection. However, the integration of nanoscale emitters into the metasurface architecture may offer an avenue to overcome this limitation and to realize metasurface able to directly emit tailored light fields. By positioning spectrally matched nanoemitters near or inside the nanoresonators, light emission processes, including spontaneous emission and nonlinear frequency generation, can be enhanced into particular spatial and/or spatial modes.

This talk will give an overview of our recent advances in the experimental realization of such light-emitting semiconductor metasurfaces.

On the one hand, we explore different strategies to integrate emitters into the metasurface, including quantum dots, monolayers of transition metal dichalcogenides, trivalent lanthanide ions, and defects and impurities in glass [2,3]. We show that a suitably designed metasurface allows for systematic reshaping of the emission spectrum and pattern, resulting in enhanced directional emission out of the substrate plane [2]. Furthermore, we demonstrate dynamic control of the emission spectra by integrating the active metasurface into a liquid crystal cell [3]. On the other hand, we study nonlinear frequency generation in metasurfaces composed of III-V semiconductors [4], offering strong second order nonlinear susceptibility. We show that the generated second harmonic is predominantly emitted into the first diffraction orders of the periodic arrangement. Furthermore, we reveal that the generated nonlinear light field depends sensitively on the metasurface geometry, the type of the resonance excited by the fundamental harmonic field, and the incident polarization. Our results indicate that resonant semiconductor metasurfaces are interesting candidates for the realization of flat sources of spatially and spectrally tailored light fields for applications e.g. in displays and lighting.

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Ultrafast magneto-elastic interactions at the nano-scale

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In functional magnetic nanostructures the acoustic, magnetic and plasmonic excitations can co-exist and interact on the nanometer spatial and ultrafast time scales. Optical spectroscopy with femtosecond laser pulses highlights a variety of nontrivial spatio-temporal dynamics, which are not only used to monitor individual excitations in real time, but also study interaction mechanisms between them, often observed in frequency mixing phenomena. In acoustics and magnetism they are often dominated by parametric resonances, where system parameters are modulated at frequencies comparable to the natural oscillation frequencies, typically in the MHz-GHz range. Here we review some experimental and theoretical investigations of magnetization dynamics driven by quasi-monochromatic and ultrashort acoustic pulses.

In recent experiments [1] the magnetization in a Ni/glass sample is excited by two distinct transient surface acoustic waves (denoted as SAW and SSLW, respectively). Magnetic tuning of the FMR frequency in resonance to their SHG, sum- and difference frequencies demonstrates the full variety of frequency mixing phenomena [2]. In contrast to nonlinear optics, the frequency mixing is dominated by the parametric effect in the externally driven FMR oscillator. An analytical model based on the resonant enhancement of frequency-mixed signals explains the experimental observations [2]. The detailed understanding of magneto-interactions in nickel thin films motivates realistic concepts for acoustic magnetization switching at the nano-scale. For example, we have performed realistic modeling for nickel nanostructures to show that an elliptical nickel nanoparticle placed in a weak external magnetic field can be switched with small-amplitude pulses of surface acoustic waves [3].

Extending our theoretical modeling beyond the acoustically-driven FMR precession allows us to study intriguing interactions between ultrashort acoustic pulses and standing spin waves (exchange magnons) in thin ferromagnetic films [4].

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The role of surface plasmon polaritons in laser processing and heating of solids

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Excitation of materials with ultrashort laser pulses induces transient changes of optical properties such as reflectivity and transmissivity. This can lead to the excitation of electrons and at some conditions Surface Plasmon Polaritons (SPP) [1,2], i.e. surface plasmons coupled to a laser-electromagnetic wave. The interference of the incident and the SPP electromagnetic waves leads to enhancement of the resulting field and, therefore, a spatial modulation of the deposited laser energy. Conditions of the SPP excitation and influence of the polarization and wavelength of the laser field on the resulting structured heating of gold are analysed in detail. We achieve this goal by developing an extended two-temperature model (TTM) [3], which takes into account the interaction of hot electrons with an additional plasmonic subsystem. Future work will focus on implementation of the SPP waves into the combined atomistic-continuum molecular dynamic-TTM model [4].

These newly developed methods for calculating optical properties and materials' heating after ultrashort laser irradiation allow us to study the fundamental mechanisms of laser energy absorption under controlled conditions. They can also be used to study the morphological effects and nanostructuring for many upcoming technological applications.

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Silicon-based nanomaterials for biophotonics

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Nanomaterials based on chemically pure Silicon (Si) are known to be biocompatible, biodegradable and possess photoluminescence (PL) in the visible and middle-IR ranges. We have studied Si-based nanocrystals (NCs), porous nanoparticles (PNPs) and nanowires (NWs) prepared by chemical, electrochemical and laser-assisted methods. The prepared nanomaterials were examined as nontoxic fluorescent labels and sensitizers for photodynamic and phothermal treatments both in-vitro and in-vivo. Si PNPs with efficient PL were produced by electrochemical etching of c-Si wafers in hydrofluoric acid solutions followed by mechanical milling in water. It was demonstrated an efficient uptake of PNPs by cancer cells as well as the bioimaging and phototherapy with this kind of NPs. Si NWs formed by metal-assisted chemical etching of c-Si wafers can be dispersed in water to form colloidal suspension for bioimaging. The prepared Si NWs exhibit strong optical absorption in the visible spectral range and the room temperature PL. Laser ablation in gaseous and liquid environment was used as a "green" physical alternative to prepare ultrapure size-tunable Si NCs and NPs for biophotonic applications in both the diagnosis and therapy, i.e. theranostics, of cancer.

Non-steady effects in resonant scattering of ultrashort laser pulses

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A survey of the recent results of the author in the unsteady high–Q resonant scattering of ultrashort laser pulses by nanoparticles is presented. It is shown that the unsteadiness of the scattering process may result in qualitative changes of the phenomenon both in the near field and in far field wave zones. The most attention is paid to the dynamics of the nonradiating anapole modes and dynamical Fano resonances, which are discussed in detail. Simple, analytically tractable models are proposed to describe the transient processes. Their comparison with the results of the direct numerical integration of the complete set of the Maxwell equations shows that the models exhibit high accuracy in the quantitative description of the phenomenon. The financial support from Russian Foundation for Basic Research (Grant № 17-02-00401) for the analytical study and Russian Science Foundation (Grant № 14-19-01599) for the computer simulation is acknowledged.

Periodic structure formation on dielectrics after irradiation with ultrashort pulsed lasers

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The significance of the magnitude of Prandtl number of a fluid in the propagation direction of induced convection rolls is elucidated. Specifically, we report on the physical mechanism to account for the formation and orientation of previously unexplored supra-wavelength periodic surface structures in dielectrics following melting and subsequent capillary effects induced upon irradiation with ultrashort laser pulses. Counterintuitively, it is found that such structures exhibit periodicities, which are markedly, even multiple times, higher than the laser excitation wavelength. It turns out that the extent to which the hydrothermal waves relax depends upon the laser beam energy, produced electron densities upon excitation with femtosecond pulsed lasers, magnitude of the induced initial local roll disturbances and the magnitude of the Prandtl number with direct consequences on the orientation and size of the induced structures.

Furthermore, a comparative study is performed to explore the periodic structure formation upon intense femtosecond pulsed irradiation of dielectrics with radially and azimuthally polarised beams. Laser conditions have been selected appropriately to produce excited carriers with densities below the optical breakdown threshold in order to highlight the role of phase transitions in surface modification mechanisms. The frequency of the laser induced structures is calculated based on a theoretical model that comprises estimation of electron density excitation, heat transfer, relaxation processes, and hydrodynamics-related mass transport.

It is envisaged that this elucidation may be useful for the interpretation of similar, albeit large-scale periodic or quasiperiodic structures formed in other natural systems due to thermal gradients, while it can also be of great importance for potential applications in biomimetics.

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3D-laser densification of porous glass: mechanisms and applications

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Porous silicate glasses (PG) are known to be promising materials for materials research due to the possibility to control their composition by impregnation with various additives [1]. In particular, recently proposed ways of a local control over the PG porosity open new directions for their applications. Thus, laser-induced modification of PG structure are shown to result not only in various local density changes, but also in the formation of periodic micro- and nanoscale regions, or, in the decomposition of the matrix revealing the formation of microcavities [2, 3].

The aim of the present report is to study the mechanisms involved in the interaction of femtosecond laser pulses with PG in bulk. Here, two main mechanisms are elucidated (i) heat-driven and (ii) stress-driven one. The resulting laser-induced material modification mostly depends on laser power density (q) in the focal area. If, on one hand, q is smaller than the one required for free-electron plasma formation in the glass, densification is observed as a result of the following thermal-elastic temporary unstable processes. – On the other hand, viscous-elastic temporary stable processes play a major role at higher q. Above the free-electron plasma appearance threshold, a local decomposition of PG structure takes place, leading to the voids appearance.

A combined laser treatment including inner densification by femtosecond laser pulses and thermal surface densification by CO_2 laser radiation was used to produce vertical walls in PG plates. Based on this result, integral devices were fabricated in the PG plate, that were found to be completely or partially impermeable to various fluids. Such approach allowed us to obtain a system of independent cells located in a single substrate plate. We note that the obtained "integrated" porous matrix can be used as a molecular barrier for separation of different molecules, or environmental indicators, when cells are isolated from each other and are impregnated with various analytes.

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Laser-structured polytetrafluoroethylene superhydrophobic surfaces as a basis of molecular transport system for sers-analyzers of ultra-small analyte concentrations

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A laser technology of fabrication of multimodal nanotexturing superhydrophobic polytetrafluoroethylene surfaces is demonstrated. The surface design with a central SERS-active target is developed, and the optimal parameters of the surface providing the most efficient transport of the analyte molecules from the solvent to the target are found.

Laser-assisted modification of Ti-6Al-4V titanium alloy surface for implantology applications

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The medicine actual problem is the biocompatible high-quality medical materials creation, as well as their surfaces processing for creating the best osseointegration conditions in implantology.

In practice, many technologies and materials for implantation are used, but they can not guarantee the successful interaction of tissues with the implant. For the implants manufacturing, titanium and its alloys are most often used, and the success of their application is directly related to the surface biocompatibility [1], which is strongly influenced by the relief and chemical composition. Also, the implant surface must be superhydrophilic for good wetting with biological fluids, which is an important property on the initial stages of osseointegration process [2].

For successful implant survival, the surface relief must be developed, must have had a large area, for better cells contact at the micro- and nanoscale [2,3]. The relief structural elements must be proportionate to biological objects. Laser processing allows the creation of such micro- and nanoscale topography, and oxidizes the titanium implants surface, increasing its wear resistance and biocompatibility [4].

In this paper, the possibility of using laser structuring and oxidation technologies for creating a variety of biocompatible structures on a Ti-6Al-4V titanium alloy is shown. The possibility of creation superhydrophilic titanium surfaces with a developed relief, which are necessary for better cells adhesion on titanium, is demonstrated. The parameters of roughness, structural profiles and the contact angle of surfaces wetting before and after laser structuring are measured. Distilled water was used as a test liquid. For overlapping pulses of more than 80%, a sharp increase in the roughness parameters Sa up to 5 microns, Sdr up to 80% and decrease of the wetting angle up to 0 degrees are observed. Cell in vitro studies have been done and have been demonstrated the positive cell adhesion response on the laser-modified titanium alloy samples surfaces.

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Laser Direct Printing of nanomaterials for flexible electronic components and sensors

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Laser Direct Printing of functional materials has emerged as a reliable, high resolution and versatile fabrication tool for flexible electronics, sensors and optoelectronic components. Current technological trends require the precise deposition of highly resolved features, in a direct writing approach which preserves their structural and electronic properties upon transfer, while increasing the number of components that can be integrated in a single device. Laser Induced Forward Transfer meets these requirements. Examples of selected applications, including organic thin-film transistors, flexible conductive electrodes comprising metallic nanoparticles and nanowire networks, and sensors based on 2D materials will be presented in this paper. Focus will be given on the Laser Direct Transfer of micrometer sized pixels of Ag Nanowire networks for the fabrication of flexible transparent conductive electrodes: low sheet resistance (< 50 Ohm/sq) and high visible light transparency ($>90\%$) make the controllable transfer of this nanomaterial highly desirable for a variety of applications. We will also discuss how Laser transfer technology can be applied for the intact and defect -free transfer of pixels of ultra-thin 2D materials. e.g Graphene enabling a whole new branch of emerging applications in optoeletronics and sensors.

Moreover, this paper will cover the application of Laser printing for the direct transfer of metal nanoparticles on polymeric substrates for the development of flexible plasmonic resonators. It has been shown that the size of the transferred metal droplets is directly related to the volume of laser-molten material region and can be controlled by the laser beam spot size and film thickness. Controllable transfer of different sizes of metallic nanodroplets will be demonstrated. The printed nanostructures have diameters from 150-500 nm, high surface ordering and reproducibility and is very promising for applications in mocochromatic filters and reflectors with tunable optical response.

3D-laser densification of porous glass: mechanisms and applications

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Porous silicate glasses (PG) are known to be promising materials for materials research due to the possibility to control their composition by impregnation with various additives [1]. In particular, recently proposed ways of a local control over the PG porosity open new directions for their applications. Thus, laser-induced modification of PG structure are shown to result not only in various local density changes, but also in the formation of periodic micro- and nanoscale regions, or, in the decomposition of the matrix revealing the formation of microcavities [2,3].

The aim of the present report is to study the mechanisms involved in the interaction of femtosecond laser pulses with PG in bulk. Here, two main mechanisms are elucidated (i) heat-driven and (ii) stress-driven one. The resulting laser-induced material modification mostly depends on laser power density (q) in the focal area. If, on one hand, q is smaller than the one required for free-electron plasma formation in the glass, densification is observed as a result of the following thermal-elastic temporary unstable processes. – On the other hand, viscous-elastic temporary stable processes play a major role at higher q. Above the free-electron plasma appearance threshold, a local decomposition of PG structure takes place, leading to the voids appearance.

A combined laser treatment including inner densification by femtosecond laser pulses and thermal surface densification by CO_2 laser radiation was used to produce vertical walls in PG plates. Based on this result, integral devices were fabricated in the PG plate, that were found to be completely or partially impermeable to various fluids. Such approach allowed us to obtain a system of independent cells located in a single substrate plate. We note that the obtained "integrated" porous matrix can be used as a molecular barrier for separation of different molecules, or environmental indicators, when cells are isolated from each other and are impregnated with various analytes.

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Laser-assisted generation of elongated Au nanoparticles and analysis of their morphology under pulsed irradiation in water and $CaCl_2$ solutions

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Laser generation of Au elongated nanoparticles (NPs) and their successive fragmentation and agglomeration are experimentally studied for the first time. Previously it was shown that laser-generation of elongated Au NPs was observed by ablation of Au colloidal solutions either with beta-active components or applied external field. In the present work first, laser-assisted formation of Au elongated nanoparticles by ablation of a solid Au (99,99%) target in water was done using a fiber ytterbium laser sources with pulse duration of 200 ns and pulse energy of 1 mJ. Extinction spectrum correlating with TEM images and size distribution measured by disc-measuring centrifuge of the mentioned above nanoparticles show the appearance of absorption signal in red region and near IR-spectrum that corresponds to longitudinal plasmon resonance of electrons in elongated nanoparticles. EDX spectrum of generated Au nanoparticles shows presence of a significant amount of calcium around of nanoparticles. Therefore further experiments on irradiation of the golden target with pulsed laser radiation with the same parameters were carried out, but already with adding a different concentration of $CaCl_2$ to the initial solution. In this case, it is also possible to synthetize elongated gold NPs, which is confirmed by extinction spectrum and TEM images.

Controllable reflection of direct-gap semiconductor metasurfaces

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Nanostructuring is a powerful tool to obtain unique, useful, and new properties of the initial bulk materials. The dielectric materials are promising for nanophotonics due to low losses in contrast to metallic.

We experimentally demonstrate laser pulse reflection modulation in the subwavelength direct-gap semiconductor structures supporting resonant behavior with Mie-type modes. Samples are periodic arrays of GaAs nanocylinders fabricated on a bulk GaAs substrate with intermediate nanocylinders between them for a higher contrast of refractive index. Nanodisks were fabricated using a recently reported procedure [1,2]. The height of the nanodisks is 300 nm, the diameter of the nanodisks varies from 200 to 350 nm, and the diameter-to-pitch ratio is 0.4 or 0.5. The reflectance spectra of the samples have peaks in the near infrared with specific fields for electric and magnetic dipole resonance.

Using I-scan and transient reflectance spectroscopy, we measured the reflectance modulation of GaAs nanodisk above the band edge of GaAs ($\lambda \sim$870 nm) and below, respectively. The pump wavelength λ=800 nm was taken for transient reflectance spectroscopy and λ=830 nm for I-scan measurements. The pump radiation generates electron-hole pairs due to single photon absorption processes. The free carriers influence on the complex refractive index, thus changing the real part of the refractive index below the band edge and furthermore the extinction coefficient above the band edge. As a result, the reflection of the metasurfaces modifies with the time due to carries relaxation and input intensity. These changes were detected by transient reflectance spectroscopy using the supercontinuum probe pulse when the pump wavelength λ=800 nm and by I-scan measurements for λ= 830 nm. The GaAs samples supporting Mie-type resonances in the range of 800 nm to 1050 nm and show the enhanced modulation by order of magnitude greater than the substrate.

We demonstrate the efficient reflectance modulation of up to 90% on the picosecond scale in the vicinity of the magnetic dipole resonance below the band edge of GaAs at a low pump fluence under 400 μJ/cm^2 and the reflectance modulation determined by absorption changes up to 30% above the band edge at a low pump fluence under 200 μJ/cm^2. There are both positive and negative changes in refractive index and regimes of modulation.

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Section 4: Femtosecond non-linear optics. Filamentation. High field THz generation.

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Scope
Self-compression, self-focusing and collapse
Terahertz science
Filamentation in various media
High-field nonlinear phenomena

Experimental study of guided discharge initiated by femtosecond laser filamentation having 10-100 cm length and -ms scale duration

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Generally, the duration of a guided discharge stimulated by femtosecond laser filamentation is defined by the high voltage source. In our guided discharge experiments a Marx generator was used for creation up to 180-kV pulse and a Tesla Coil for up to 360-kV pulse. More exactly, the discharge duration is defined by the product of the Marx "stack" capacitance or the Tesla geometrical output one and there load resistance. Both a circuit resistance and any discharge resistance present the load charge. In both cases the discharge duration was inside 1 mks.

For different applications a guided discharge of much longer duration is required.

We present here experiments carried out with the discharge length from 8.5 cm (Marx generator) up to 100 cm length by using the Tesla coil HV source. To increase the duration of the discharge we employed a second circuit which injects an additional current pulse under much smaller voltage. Our first experiments with prolongation of 85-mm discharge up to 130 μs were presented in [1]. With help of high speed imagery we showed that 10-100-cm discharge guidance has been achieved successfully during more than 1 ms. Different characteristics of the discharge are presented.

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Two-color femtosecond soliton bound states

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In [1,2] we demonstrated the high potential to manipulate the dynamics and properties of ultrashort light pulses by group-velocity matched interaction in nonlinear dispersive media. The scheme can be used for the generation of highly coherent supercontinuum spectra, or the efficient compression of pulses into the few-cycle regime. Here we transfer the mechanism to the incoherent interaction between two pulses located in separated anomalous dispersion regimes. This allows for different types of interaction scenarios enabling the control of energy transfer in a versatile manner. Besides regimes of trapping and fusion processes, the generation of two-color molecule states possessing a nonzero binding energy can be realized. The bound state thereby can consist of only a few optical cycles and exhibit a high density over a broad spectral range. We demonstrate the robustness of the molecule states, and the possibility to evaporate soliton states from the bound states.

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Suitable input conditions for femtosecond pulse tight focusing into dense medium

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Despite the significant progress in the simulations in femtosecond optics [1], the simulations of high-power laser beam propagation in nonlinear medium in experimental conditions of ∼1-cm-wide beam focused by a ∼20-cm lens still is a challenging task in terms of computational resources. The optical breakdown in air provide the ∼100-μm-wide plasma channels, so for the accurate numerical modelling one should have a transverse numerical grid with about 1 μm step. Still it is possible, if the time resolution is not an issue for the physical task, i.e. in stationary approximation [2,3]. Whether the carrier-wave-resolved simulations are required (particularly, for the studies of generation of high harmonics or terahertz radiation), the total rapid access memory required for data array at a certain distance z approaches terabytes.

The procedure of suitable input conditions (SIC) [4] construction is beneficiary for this task. The typical parameters of the air-based on-table experiments allows the choice of the intermediate plane $z = z_0$ between the focusing optics and the waist so that the nonlinearities can be neglected in propagation before this plane. This is ensured by the negligible plasma density at the point and the minor value of B-integral on the path before z_0. Both this values can be estimated analytically. For the parameters of the experiment, i.e. $a_0 = 0.5$ cm, $f = 15$ cm, $W = 1.5$ mJ, $t_0 = 50$ fs, the SIC-plane can be placed very close to the focal plane, at $z_0 = 14$ cm.

Thus, provided with the field incident to the lens, we can linearly propagate each Fourier harmonic of the field to z_0 using Forward Maxwell equation, which describe accurately even the focusing with NA ≈ 0.6 [5]. It is notable that FME can be solved analytically for the input representing the Hermite-Gaussian mode. Otherwise, this routine requires about $2^{15} \times 2^{15}$ points for non-axially-symmetric centimeter-wide beam, as we need a micrometer resolution. After propagation to z_0, we extract only the central part of the numerical grids, where the squeezed beam appear. Typicaly, it will be $2^{11} \times 2^{11}$ points for each harmonic, as for the chosen geometry the beam shrinks by 16 times at z0. Thus, the overall array for 4D propagation will be $2^{11}(x) \times 2^{11}(y) \times 2^{12--14}(t)$ points.

We apply the aforementioned procedure for the simulations of THz generation from the chirped pulses in (t, r, z) geometry and for the simulations of the third harmonic generation from the regularized beam in (t, x, y, z) geometry.

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THz generation by two-color filamentation at different wavelengths

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In the electromagnetic spectrum, the terahertz (THz) frequency range is located in-between microwave and far-infrared frequencies. For many reasons THz radiation attracts a lot of attention [1,2]: large biological molecules have structural resonances at THz frequencies; THz devices could play a role of a bridge between high frequency electronics and photonics; THz radiation has high penetration depth in many materials, but unlike x-rays it does not ionize them.

Despite the high potential of THz radiation for applications, there is still a huge lack of powerful THz sources. Nowadays, the most powerful table-top sources of THz radiation are based on optical rectification in nonlinear crystals or two-color filamentation [3,4]. In our work we are interested in the two-color filamentation technique, since it allows one to generate single cycle THz pulses with high peak power and opens a way for nonlinear interactions of THz radiation with matter.

Despite of the existing experimental and theoretical studies [5-7], there is still an open question about the optimal laser source for the two-color filamentation technique. To answer this question, we use numerical simulations to study the generation of THz radiation by two-color filamentation of laser pulses with different wavelengths. We consider the wavelengths in the range from 0.6 to 10.6 μm, thus covering the whole range of existing or possible in the future laser sources. We show how the parameters of two-color filaments and generated THz pulses depend on the laser wavelength.

We demonstrate that for two-color filamentation there is an optimal wavelength of laser source that provides the highest THz conversion efficiency.

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Enhancing x-ray generation under interaction of chirped pulse induced filament with solids placed in air

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Femtosecond laser filamentation in air attracts considerable interest because of a wide range of applications such as remote spectroscopy and filament-induced laser micromachining [1,2]. In these tasks it is important to know intensity inside femtosecond laser filament. One of the methods of intensity estimation is based on signal ratio measurement of two nitrogen fluorescence lines [3]. Another one is based on the measurement of the laser energy transmitted through small diaphragms which are fabricated as a result of plasma assisted ablation by the filament itself in a thin metallic foil [4]. Based on these methods air filament intensity is estimated as $(0.3 - 1.4) \cdot 10^{14}$ W/cm^2. It can be enough for X-ray generation from laser-solid interaction [5]. It should be noted, that the efficiency of x-ray yield under interaction of femtosecond laser radiation with solid target can be enhanced using chirped pulses [6].

The main goal of this work is to investigate x-ray yield from microplasma, produced under ablation of metal (Fe, Cu) target by femtosecond filament in the regime of transform-limited and chirped pulses; to estimate intensity inside filament using data from x-ray spectra.

We found that interaction of femtosecond Ti:Sa laser in the filamentation regime (lens focal length F=100 cm, laser power is 5 times larger critical power $P \sim 5P_{\mathrm{cr}}$) during micromachining is accompanied by production of x-ray photons with energy more than 3 keV. We expect that increase of pulse duration (using chirped pulses) in monofilament mode can lead to increase of x-ray yield. It was obtained that x-ray yield increase up to 4 times using positive chirped laser pulses (∼500 fs) in compare with transform limited (∼60 fs) pulses at laser power of about $P \sim 5P_{\mathrm{cr}}$ in monofilamentation regime. The rise of x-ray yield is also accompanied with rise of intensity of emission lines and ablation rate and up to 10 times and enhancing length of filament.

We measured X-ray emission spectrum from plasma created under filament-target interaction in regime of positively chirped pulses and deduced the hot electron temperature from it, which is about T_h∼1,5 keV (fig.1).

Using link between T_h and intensity I [5]: $T_{\mathrm{h}} = (5.3 \pm 0.2) \cdot (I\lambda^2)^{1/3}$ we found that intensity in filament is about $I = (2,0 \pm 0,3) \cdot 10^{14}$ W/cm^2.

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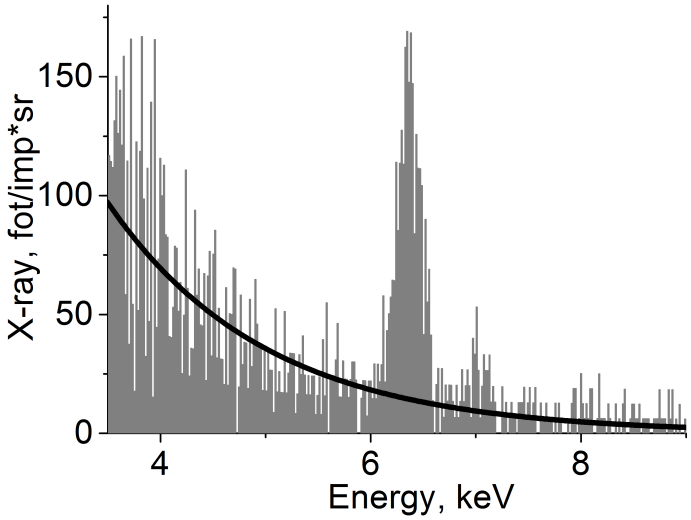

Figure 2: X-ray emission spectra of iron in range from 4 to 9 keV using chirped laser pulses (500 fs). Line represents a fit of bremsstrahlung background and indicates a hot electron temperature of T_h ∼1,5 keV.

Nonlinear-optical anisotropy in silicon nanowire ensembles

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Recent searches for novel media for photonics led scientists to artificial media formed by dielectric wavelength-scale particles. A special interest is paid to metamaterials based on high-index media since they allows resonances and concentration of the field inside the nanoparticles. However, the most of studies are focused on the nanowires of submicrometer length. Meanwhile, increase of the nanowire array thickness can result in efficient light scattering, which, in its turn can increase the efficiency of many optical processes, including drastic change of total reflectance and enhancement of optical process efficiency.

Arrays of the silicon nanowires (SiNW) of about hundred nanometers in diameter and of 1 to 100μm in length are examples of these structures. Due to strong light scattering in them SiNW arrays demonstrate enhanced total reflection in transparency spectral region combined with huge (above 95%) absorption of visible light. Moreover, in comparison with crystalline silicon (c-Si) SiNW arrays exhibit increased efficiencies of interband photoluminescence, Raman scattering, second- and third-harmonic generation, coherent anti-Stokes Raman scattering, and the light self-action. The light propagation in SiNW array was found to be diffuse-like. These facts open up new possibility for formation of silicon-based solar cells of enhanced efficiency, all-optical sensors and nonlinear-optical devices.

The SiNWs were formed by means of metal-assisted chemical etching of (110) c-Si wafer. The SiNW were tilted at 45° to the wafer surface. To characterize SiNW ensemble nonlinear-optical anisotropy we registered the third-harmonic (TH) pumped by Cr:forsterite laser radiation (1250 nm, 80 fs, 80 MHz, 2 nJ). Despite strong light scattering in the SiNW ensembles, significant nonlinear-optical anisotropy of the TH signal was found in them. TH orientation dependence of the signal differs from one for c-Si and depend on incidence of the exciting radiation (along the SiNWs or perpendicular to them). The latter factor significantly influence TH signal magnitude. The obtained effects are connected with interplay of light scattering in the SiNW ensemble and enhancement and anisotropy of the local field inside SiNWs.

Multifrequency stimulated raman scattering in condensed media under ultrashort laser pulses excitation

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Stimulated Raman Scattering (SRS) in condensed media has been studied before with using of nanosecond laser pulses as exciting emission. At high enough exciting light intensity Multifrequency Stimulated Raman Scattering (MSRS) has been observed in different substances: water, benzene, CS_2, $CaCO_3$ and others. However, using of nanosecond giant laser pulses as exciting emission with very high laser pulses energy results in the destruction of condensed dielectric media and restricts MSRS investigations. So the using of ultrashort laser pulses as exciting emission with small enough energy pulses, but very high density of laser power for MSRS investigations of crystals and liquids is preferable. In this paper the experimental results on MSRS spectra investigations on the base of ultrashort laser pulses as exciting emission with wavelength 532 and 1064 nm of Nd:YAG laser are presented. Solid state Nd:YAG laser generated 80 ps pulses at 1064 nm, and 60 ps pulses at 532 nm.. We have observed Stokes and anti-Stokes Raman satellites in MSRS spectra for a number of substances: liquids (light and heavy water, ethanol, glycerin) and crystals ($CaCO_3$, $NaBrO_3$, $Ba(NO_3)_2$, $KGdWO_4$, LiOH, LiOD). The different physical mechanisms of MSRS in water has been studied. The important property of heavy water is that it does not absorb the neutrons. Therefore, heavy water is widely used in the nuclear reactors as the substance for the retardation of neutron beams velocity, and has the great importance for realizing thermonuclear synthesis. In this paper, we present the results of MSRS studies in light and heavy water excited with picosecond laser pulses at 532 and 1064 nm. Raman thresholds in light and heavy water, and SRS efficiency versus the pump pulse energy have been measured. Near Raman threshold, Stokes components were observed with spectral lines which are not split. At increasing the intensity of laser exciting radiation, the physical conditions in water are changed. In water, laser-induced plasma and shock waves of pressure propagating with supersonic speed are formed. The number of sub-components increases with increasing radiation intensity. Thus, with picosecond MSRS in water, a transient phase transition of water into various forms of ice is possible. The onset of plasma formation can be determined by the beginning of the splitting of the Stokes components. A lot of Stokes and anti-Stokes Raman satellites has been recorded in MSRS spectra of $Ba(NO_3)_2$ crystals. In the case of 532 nm Nd:YAG laser excitation such satellites formed frequency comb emission in visible and violet spectrum range. In the case of 1064 nm excitation a number of Stokes and anti-Stokes Raman satellites have been generated in infrared and visible ranges. Thus MSRS in liquid and solid media opens the opportunity to realize the frequency comb laser generation in wide spectral region.

Four-photon absorption measurements in fused silica at 480 nm

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The four-photon absorption cross-section in fused silica KU-1 at a wavelength of 480 nm was obtained for the first time by measuring the dependence of the sample transmission on the intensity. For the measurement, we used a thin (0.1-mm-thick) sample to minimize the B integral, which did not exceed 3 in our experiments (we measured in a separate scheme the nonlinear refractive index for our sample, the value was $n_2 = (2.5 \pm 0.3) \cdot 10^{16}\text{cm}^2/\text{W}$). The transmission was measured as the ratio of the input and output energies at different intensities measured by CCD camera. For this purpose, we used two techniques: I-scan (the incident energy was varied at a constant position of the sample) and Z-scan (the sample was moved along the converging beam at the constant incident energy). Scheme of the experiment is shown in fig 1.

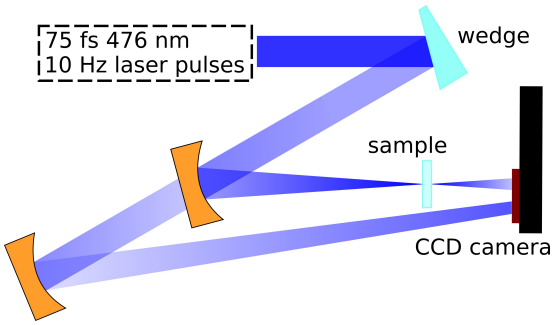

Figure 1: Scheme of the measurement of the four-photon absorption coefficient.

The figure 2-3 show the transmission curves normalized to transmittances at low intensities for these two cases.

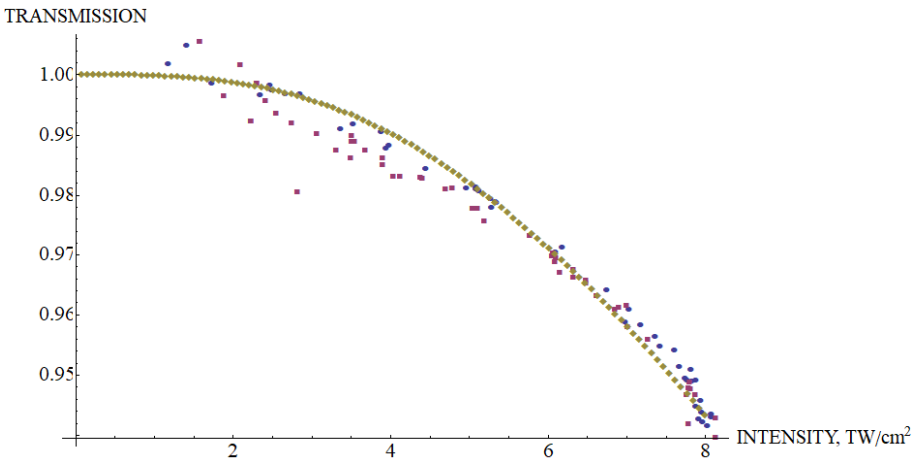

Figure 2: Normalized transmission of the sample against incident intensity (I-scan). The brown curve is the theoretically obtained dependence for the absorption coefficient of 0.13 $\text{TW}^{-3}\text{cm}^5$.

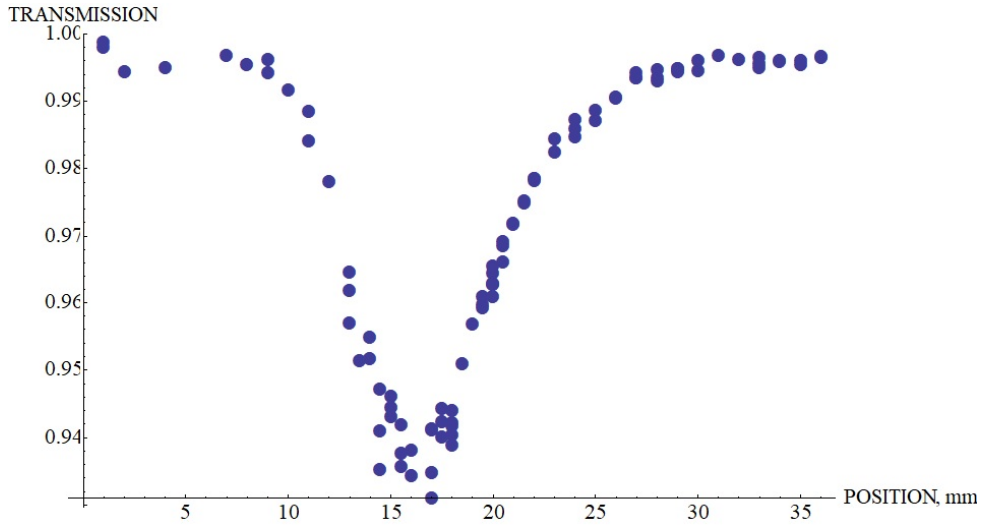

Figure 3: The normalized transmission curve obtained by the Z-scan method. The dip corresponds to the absorption coefficient of 0.15 $\mathrm{TW}^{-3}\mathrm{cm}^5$.

After analyzing the errors, the value (0.14 ± 0.07) $\mathrm{TW}^{-3}\mathrm{cm}^5$ was chosen from the experimental results, which corresponds to $\sigma_4 = (1.0 \pm 0.5) \cdot 10^{-115}$ $\mathrm{cm}^8\mathrm{s}^3$, which is an order of magnitude smaller than the values of the four-photon absorption cross-sections known from the literature.

Generation of terahertz radiation by two-color femtosecond ionizing pulses with arbitrary polarizations of components

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We study both analytically and numerically the dependences of terahertz (THz) yield on ellipticities and intensities of the one-color components of two-color femtosecond ionizing pulses with various frequency ratios between components: 2/1, 3/2, 4/3, 5/2, 4/1. Our results show that for the frequency ratios of the form (m+1)/m, where m is a natural number, there is a significant increase of THz yield when using properly elliptically polarized components, while for the rest (such as 5/2) linear polarization is almost optimal.

Photoacoustic imaging of femtosecond filament in water

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Laser-induced filaments in liquids and gases leads to the creation of plasma channel. The high pressures and temperatures inside the channel generate shock-waves generation and leads to the formation of cavitation bubbles[1]. The shock-wave profile can be measured in the far zone and can be used for the restoring the initial pressure distribution. This distribution is determined by the energy delivery of the femtosecond filament. Therefore can retrieve the filament profile using photoacoustic imaging [2]. In this work, we demonstrate a first-of-its-kind two-dimensional (2D) photoacoustic imaging of a single femtosecond filament in water and determine its location and size. The filament was generated by intense Cr:Forsterite femtosecond laser pulses (central wavelength 1240 nm, laser energy up to 30 μJ, pulse duration 170 fs, repetition rate 10 Hz). Acoustic signal induced by a filament was recorded by a single 100MHz-band piezoelectric detector rotating around the filament with 1° angular resolution. After filtering the experimental data and applying the back-projection reconstruction technique [3], we retrieved 2D filament-induced l pressure distribution in water.

The obtained filament size (FWHM) is about 50 μm (see Fig.1), that is in a good agreement with a literature data [4]. Additionally, using the technique of a e single-shot darkening of photographic plates we obtained that the filament size is $58 \pm 6\mu m$. It is important to mention that the photoacoustic imaging also gives opportunity to direct measurements of the filament reservoir (the red halo around the core on the fig.1).

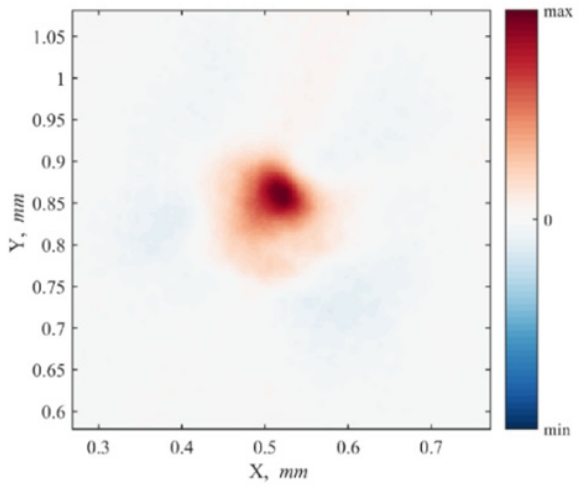

Figure 1: Retrieved 2D photoacoustic tomogram.

The optoacoustic tomography can be applied to direct measurements of the energy delivery into the medium under filamentation of ultrashort laser pulses, that other methods of filament profile measurements can not offer. In combination with relative simplicity

and high accuracy it makes optoacoustic tomography a unique instrument for filamentation investigations.

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Nonlinear optical properties of CO_2 and Xe in sub- and supercritical states: anomalous behavior of nonlinear refraction index and supercontinuum generation

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Fluid at supercritical state have an intermediate properties between gas and liquid, that makes it an unique medium with advantages of either states of matter [1]. High and adjustable nonlinear properties of supercritical fluids (SCF) make them an ideal tool for controlling the nonlinear optical processes using high power laser femtosecond laser such as filamentation , spectrum control, spatial and temporal parameters of laser radiation [2].

There are large density fluctuations, and cluster formation in SCF, which has maximum at Widom line (the line dividing supercritical region into gas-like SCF and liquid-like SCF) [1]. To characterize the nonlinear optical properties of SCF we carried out experiments with a Cr:forsterite based laser system operating at λ =1240 nm, at 10 Hz repetition rate, giving laser pulses of 170 fs and pulse energy of 30 μJ. The collimated laser beam passed through the supercritical cell with explored fluid. The broadening of the laser spectra give us opportunity to estimate the nonlinear refractive index [3]. The pressure dependence of the nonlinear refractive index is presented in Fig.1a.

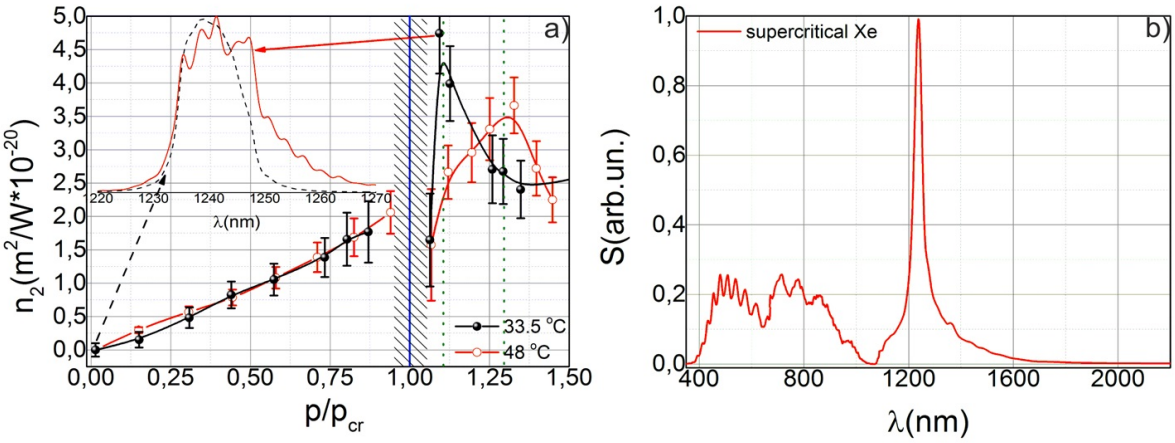

Figure 1: (a) The pressure dependence of nonlinear refractive index (n_2) for CO_2 at different temperatures. The shaded zone shows the region where n_2 measurements are not allowed to obtain reliable data. The vertical blue line shows critical pressure. Dotted lines show ridge locations. The insets show the measured spectrum of 170-fs laser pulse passed through the cell at different pressures. (b) The combination of supercontinuum and emission lines in supercritical Xe (69 bar, 25°C).

We for the first time show, that the clustering of the matter, which manifest itself maximally in the vicinity of the Widom line, leads to the anomalous increase of n_2 [3]. Under experimental conditions the high values of n_2 provides the efficient supercontinuum

generation and plasma creation accompanying filamentation. As the result, the visible wing of the spectrum is a combination of the supercontinuum and the emission lines of Xe (see Fig.1b). The obtained spectra can be precisely controlled by varying pressure inside the cell. Therefore, by simple varying the laser pulse energy and pressure one can generate adjustable spectra broadening from 400 nm up to 2 μm.

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Highly nonlinear volumetric photographic fluorescent materials

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The report will address scientific problems related to the development and application of new extremely sensitive luminescent methods for studying the interaction of intense laser radiation with matter. These methods are based on the use of the new functional solid-state media - nonlinear bulk dielectric photographic luminescent materials in combination with means and methods of luminescent micro-tomography with a time resolution. Such media proved to be very useful for studying the filamentation of laser radiation and other highly nonlinear processes of interaction of light and matter. They make it possible to experimentally study the spatial distributions of intense pulsed laser fields in media, to memorize them, to visualize and to study them by highly sensitive stationary methods. The experiments were carried out with single-crystal and ceramic media based on wide-band alkaline-halide and alkaline-earth compounds. Their interaction with femtosecond laser pulses of titanium-sapphire and ytterbium lasers was investigated. Photosensitivity of such crystals is due to the formation under the action of intense laser radiation of stable structural defects that are capable of luminescence with additional optical excitation. Intensive laser radiation is absorbed by the electronic subsystem of the crystal and ensures the generation of electron-hole pairs. Luminescent defects are formed in the nuclear-core subsystem. The connection between the subsystems, which leads to defect formation, is provided by anionic excitons, which arise when recombination of electron-hole pairs. Then the excitons spontaneously decay into pairs of Frenkel defects, the components of which are mobile at room temperature. They undergo recharging, migration, and aggregation. As a result, stable color centers that luminesce upon photoexcitation with high yield are formed. Spatial distributions of the intensity, spectra, and duration of the luminescence of defects induced by interaction with laser radiation were studied using a confocal scanning luminescence microscope with a time resolution of the MicroTime 200 operating in the photon counting mode. It is established that the efficiency of formation of simple and aggregate color centers in cubic LiF crystals at a normal incidence of the laser beam on the plane of the cube face periodically depends on the azimuth angle θ between the electric vector and the edge of the cube on this face with a period of $\pi/2$. It is found that the azimuthal dependences, for defect formation (maximum at $\theta = \pi/4$) and for photogeneration of carriers (maximum at $\theta = 0$) are in antiphase. The calculations have shown that the self-focusing and filamentation processes controlled by the components of the third-order nonlinear susceptibility tensor are most effective for the orientation $\theta = \pi/4$. The experiment showed that with this orientation, the critical power and self-focusing length are reduced, so that the density of the filaments increases and, as a result, the average concentration of color centers created by laser filaments increases. It is known that multiple filamentation is stimulated by inhomogeneities contained either in the laser beam itself or in the medium with which the beam interacts. A luminescent method for detecting inhomogeneities is proposed, which play a key role in the mechanism of multiple filamentation in the highly nonlinear interaction of light and matter. The method includes a line scan of the sample by a periodic sequence of femtosecond laser pulses in

the filamentation and defect formation regime. Multiple filamentation in interaction with the photosensitive media of coherent pairs of shifted femtosecond pulses running along the same path with different velocities was investigated. Methods are developed for longitudinal positioning of the action of light on the medium by means of coherent pairs of pulses. Technologies for recording monochrome and full-color images in optical media based on dielectric crystals and ceramics have been developed and tested.

The influence of air humidity on the ultrashort pulses filamentation

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The concentration of water vapor in ambient air is a variable value and typically it doesn't exceed several percent. Its influence on the UV, visible and near IR emission propagation is usually negligible. However, in [1] relative humidity growth from 0% to almost 100% led to the tenfold increase in the density of plasma generated by 248 nm wavelength emission. As ionization rate strongly influences the parameters of filaments, the filamentation phenomenon can also be affected by the humidity change. Therefore, our goal was to investigate the linear plasma density distribution and the energy deposition at different relative humidity values of air under the filamentation of UV (248 nm) pulses in comparison with IR ones.

In the experiments we used the pulses of the first and the third harmonics of Ti:Sapphire laser system with central wavelengths of 744 nm and 248 nm correspondingly. The beam was focused to a chamber with a lens f=100cm placed on a rail. We measured linear plasma density along the filament in dependence on distance from the lens by means of a system of electrodes [1]. The measurements were conducted for three levels of relative humidity (RH) of air: 0%, 30% and 90%.

We observed no significant difference in the parameters of plasma channels generated under filamentation of IR laser pulses at different RH. In the case of UV pulses the amplitude of linear plasma density was almost one order of magnitude higher for RH=100% than for RH=0%, the length of plasma channel doubled with RH increase. The maximal energy deposition in the case of filamentation of UV pulses in a moist air was obtained to be approximately a quarter of the initial energy.

The numerical simulation of UV pulses propagation in filamentation regime in air at different relative humidity values were conducted with the pulse parameters similar to the experimental ones; the cross-section of water vapor photoionization was taken from [1]. The results of computer simulation were in good agreement with the experimentally obtained ones.

Thus, the concentration of water vapor was shown to significantly influence the parameters of plasma channels formed under the filamentation of UV pulses unlike the case of IR emission filamentation. High ionization efficiency in humid air also leads to the considerable energy deposition. This investigation was supported by Russian Foundation for Basic Research (grant № 18-32-00726).

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Ionization and explosion of a spherical water droplet in air by femtosecond laser radiation at intensities of the order of magnitude achieved at laser filamentation

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The interaction of femtosecond Ti:Sa laser radiation beam with high incident intensity which was enough for filamentation in ambient air and water droplets with size less than the filament core was studied. Earlier the explosion of water droplets due to high intensity femtosecond radiation was studied. The dependence of explosion character on the incident intensity was studied [1], also the scattering of laser radiation and its spectral changes was studied in [2]. Nevertheless the interaction at intensities as higher than 10^{14} W/cm^2 which are supposed to be reached at the filament core [3] has not been studied yet. We observed experimentally the interaction and the explosion of 30 μm in diameter water droplet with laser radiation at the position of it filamentation. The hot plasma white light spots was observed at back and front foci posed inside the droplet and even in the air lying behind the droplet. The surface of droplet is also ionized.

The laser matter interaction and energy absorption was studied numerically using the self-consistent model, directly solving Maxwell equations coupled with a balance equation for electron density.

The research is supported by the RFBR (grant № 18-32-00949).

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The laser plasma filament in the air visualization by the nonlinear phase contrast imaging using Kerr nonlinearity of the fused silica

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The phase contrast method provides an opportunity of imaging transparent (phase) objects without implementation of traditional interferometrical techniques. The obtained images do not need to be processed after handling as, for example, fringes in the interferogramms. The nonlinear phase contrast has an advantage over traditional phase contrast microscopy due to simple way of its optical scheme alignment. Time resolution is determined by the type of nonlinearity implemented in nonlinear imaging scheme.

In this research the nonlinear phase contrast method was used to obtain an image of laser plasma. The laser radiation was divided to two parts, the most powerful to produce plasma using and the second part used to probe this plasma. The 1 mm thick fused silica was settled at the focus of the imaging system to obtain the nonlinear phase shift required for nonlinear imaging. The intensity of the probe beam was sufficient to provide enough value of the B-integral for the central part of the probe beam. The phase contrast images of the laser plasma was obtained for the first time in the scheme of nonlinear phase contrast imaging using Kerr nonlinearity of the fused silica. Almost instant response of the fused silica Kerr nonlinearity lead to high time resolution and gives an opportunity to investigation of fast processes as laser plasma filamentation.

The research is supported by the RFBR (grant № 18-32-00949).

Geometric-optics correction of initial conditions for non-paraxial propagation equations

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Propagation equations are widely utilized for the theoretical description of laser-matter interaction processes, such as high harmonics and supercontinuum generation, filamentation etc. due to the natural separation of the evolution or longitudinal coordinate from transverse coordinates (and time) [1]. So, the application of propagation equations to the extreme focusing conditions remains a challenging task, because the high-numerical-aperture (NA) optical element results in the notable coupling between the evolution coordinate and the others. Unidirectional pulse propagation equation [1] describes the nonlinear propagation of radiation with arbitrarily wide spectrum, temporal and spatial. This equation recommended itself in the best way for the THz generation, which diverges at large angles of 45° and even more [2].

Recently, we showed that the application of non-paraxial propagation equations to the extreme focusing ($f/1$) is limited not by equation itself, but by the chosen form of initial conditions [3]. In [3], the suitable input condition paradigm was first pronounced: one should obtain the field distribution in the plane laying between focus and focusing element, using, e.g., diffraction integrals, the exact integral solution of Maxwell equation. This routine repairs the solution of propagation equation, though requires significant numerical efforts itself. Later, the methods of transformation optics were used [4] to successfully avoid the need for suitable input. This work established the unexpectedly high validity of the paraxial propagation equations: "classical" input conditions worked satisfactorily for the high–NA objective up to 0.6.

In this work, we build the in-plane input conditions for nonparaxial propagation equations by means of geometric optics. We consider the large angles of reflection and the corresponding wavefront delays with respect to the initial plane. As a result, the amplitude and phase of the input beam are modified as compared with the "classical" input. The numerical simulations with scalar UPPE demonstrated the excellent agreement with the solution by diffraction integral [5] up to NA = 0.6 ($f\# = 0.6$) for the different reflecting focusing elements (i.e. $\phi \sim r^{\alpha}$; $1 \leq \alpha \leq 2$, from conical to parabolic reflecting surface). This procedure is numerically fast, as it requires only the solution of algebraic equation for every point in the input plane.

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Acoustic signal for femtosecond filament plasma grating characterization in air

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Diagnostics of high-intensity zone produced by ultrashort laser pulse filamentation [1] is a nontrivial task requiring specially designed setup, since almost any material inserted into a high-intensity beam will be destroyed. In parallel with filament study by optical methods, photoacoustic methods were employed to filament characterization [2]. The sensitivity of photoacoustic method is enough to detect the energy deposition into air in the prefilamentation regime without plasma generation. High-resolution imaging interferometry technique [3] may be complemented by straightforward measurement of air density variation using wideband piezoelectric transducers [4]. Thus, for the past 4 years the acoustic method in studying single and multiple femtosecond filaments has advanced significantly. It was shown that broadband acoustic detectors can be used to noninvasively analyze both energy deposition and fine structure from plasma filaments.

In this paper we researched numerically the acoustic waves emission from dynamic plasma grating produced by the interference of two noncollinearly propagating beams from the same pulse [5]. The origin of the acoustic wave is the air density perturbation after the plasma recombination in the grating. The directional diagram and frequency spectrum of the acoustic wave induced by the grating are studied and two characteristic spectral maxima corresponding to the overall plasma width in the filament and the width of one strip of the grating (the grating period) are identified numerically based on the solution of the full wave equation for acoustic signal propagation [6].

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Harmonic generation in mid-infrared laser filaments in gases

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I will discuss experiments on the generation of supercontinuum and low-order harmonics in femtosecond laser filaments in gases. Two different laser sources have been used: A carrier-envelope-phase (CEP) stable, 11 fs OPCPA at 1.7 μm wavelength at the University of Central Florida (the group of Prof. Z. Chang) and a CEP-unstable, mid-infrared OPCPA at 3.9 μm wavelength at the Technical University of Vienna (the group of Prof. A. Baltuska). In both experiments, we observe CEP-dependent interference in the spectra of white light generated through filamentation of infrared laser pulses in air and argon. We interpret these spectral features as being the result of interference between different nonlinear source terms generating low-order harmonics and supercontinuum. We suggest that the interference effect observed in our experiments could be used for in-line, single-shot measurements of the CEP in CEP-unstable sources. Numerical simulations of our experiments quantify the contributions of different nonlinear source terms in harmonic generation.

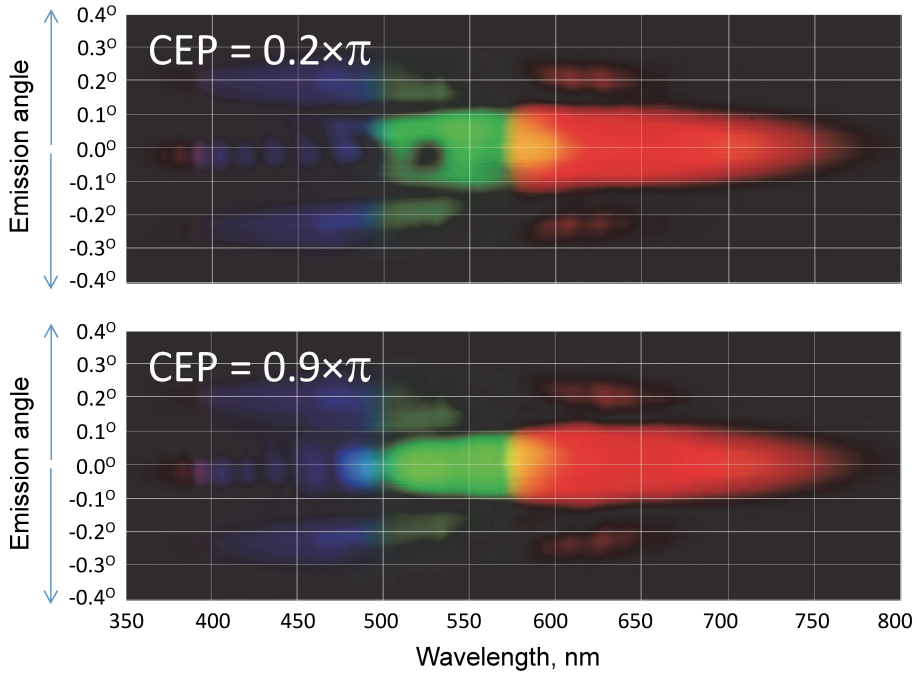

Figure 1: Experimental k – ω spectra of the forward-propagating white-light emission produced through filamentation, in the atmospheric-pressure air, of two-cycle, CEP-stable laser pulses at 1.7 μm center wavelength. The 12 mm – diameter beam is focused, in air, by an $f = 2$ m concave mirror. In the images, the wavelength of the emission is resolved along the horizontal axis, while the emission angle is resolved along the vertical axis. The appearance of the spectra is π-periodic in the CEP of the laser pulses. The two cases shown correspond to the extreme values of the CEP specified in the images (relative to a common arbitrary number), that produce the most dissimilar spectra. The spectral feature with the strongest CEP dependence is the appearance and the disappearance of the spectral hole in the on-axis emission near the wavelength of the third harmonic of the driver, at around 525 nm.

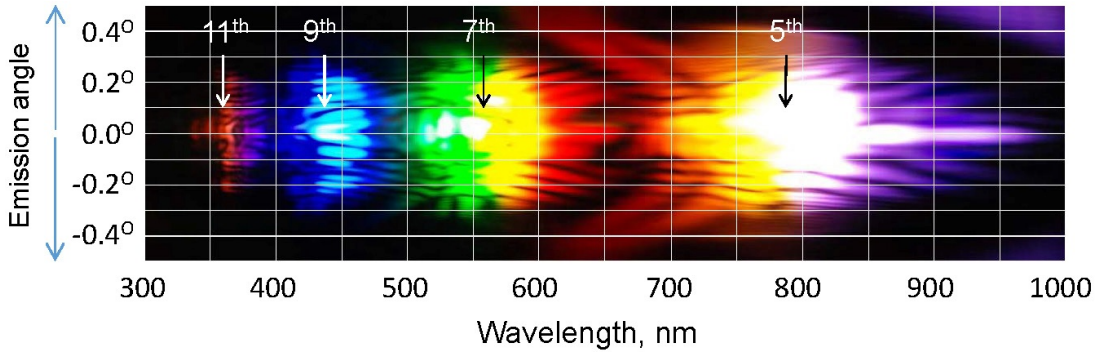

Figure 2: Angularly-resolved emission spectrum of white light generated through filamentation of an ultrafast laser pulse at 3.9 μm wavelength in air. Our detection system captures overlapping, yet distinct harmonics of orders from 5 to 11. Emission spectra generated in argon show CEP-dependence spectral interference between third and fifth harmonics of the mid-infrared optical driver.

This work has been made possible by the contributions of collaborators from the research groups of Profs. Zenghu Chang, Andrius Baltuska, Miroslav Kolesik and Dr. Arnaud Couairon. The author's contribution was supported by the US Air Force Office of Scientific Research under program FA9550-16-1-0013.

Femtosecond laser superfilamentation under various focusing conditions

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We investigate filament spatial interactions within femtosecond multifilaments in air under various focusing conditions in stochastic and regularized regimes. A bunch of filaments was formed by radiation of a Ti:Sa laser system delivering pulses with 55 fs duration and 5–25 mJ energy at 10 Hz repetition rate with 7mm FWHM beam diameter. Focusing conditions have been changing by introducing lenses with 20 cm – 3 m focal lengths into optical path. Regularized multifilament was optained by introducing amplitude mask before the lens. We showed that superfilament is formed only in the case of weak additional focusing (3 m and 1 m lenses). In this regime a small number of filaments (several times smaller than P/P_{cr} ratio) is formed. Each of them contains higher plasma concentration than a single filament under the same focusing condition. The energy deposition from each filament of this bundle is more than 1 order higher than from the single filament. Otherwise, under more tight focusing filament bunch doesn't form similar unified stuctures. Energy deposition grows linearly with the pulse energy and plasma concentration doesn't considerably exceed this value for a single filament under the same focusing conditions. We also revealed that at the energies that correspond powers above the $5P_{cr}$ superfilament tends to split into several filaments (each of them with higher plasma concentration than single filament has), so one cannot fuse filaments in a bunch into one filament with arbitrarily high plasma concentration.

Terahertz wave generation from liquied gas

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We present results of our research on generation of THz radiation in liquid nitrogen. We used a dual-frequency scheme when emissions of the main laser frequency and its second harmonic are mixed in the same medium. The research showed a possibility of effective conversion of optical radiation into THz radiation.

The source of electromagnetic radiation in the THz band on the basis of laser spark was firstly presented in [1]. An experiment in which a liquid, namely, water was used for the conversion of femtosecond radiation into the THz one is described in [2]. Water is a polar liquid which has high absorption in the THz frequency range and the authors of previously published works have to use for the experiments the very thin water films. Unlike water, considerable absorption both in THz and NIR ranges is absent in liquid nitrogen.

In our experiments the laser beam ω is directed vertically from top to bottom with the help of a set of mirrors and is focused by a lens inside the LN in Dewar vessel. The Dewar vessel is mounted on a microscopic translation stage and can be moved vertically, which enables the regulation of the position of the lens focus regarding the surface level of the LN. THz radiation generated in the beam-waist is reflected from a flat aluminum mirror and is collimated by the parabolic mirror.

First of all, we obtained the THz radiation with the use of a dual frequency scheme in an experimental set-up without liquid nitrogen, from a routine optical air breakout. After that, the laser beams on the fundamental and the second harmonic was focused into LN and the THz radiation was also observed. Fig. 1 shows the dependences of the THz pulse energy on the lens focus position regarding the surface of liquid nitrogen (the laser beam waist is located inside the liquid if $z < 0$ and in the air if $z > 0$). The intensity of the generation changes exponentially as the beam-waist position varies and a leap in the level is observed when the level of the surface is passed.

Also we have studied how THz yield scales with laser pulse duration and its energy, angle of rotation of the BBO crystal and measured spectra of the THz radiation.

The present work describes broadband generation of THz radiation first obtained in liquid gas. Unlike previous attempts to generate THz radiation in water, in our experiments we used liquid nitrogen, and the results enabled us to suggest a physical mechanism of this process.

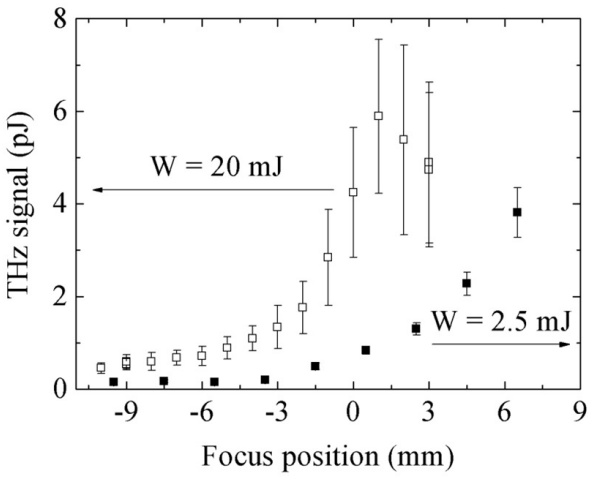

Figure 1: Dependence of THz pulse energy on the lens focus position regarding the surface level of the liquid nitrogen. "black squares" indicate THz signal with 2.5 mJ pumping energy; "open squares" indicate THz signal with 20 mJ pumping energy.

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Nonellipsometric electro-optic sampling of terahertz pulses in GaAs

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Electro-optic (EO) sampling of terahertz waveforms by femtosecond laser pulses is widely used in THz time-domain spectroscopy. In the conventional scheme of EO sampling, the probe optical pulse propagates collinearly with the terahertz pulse in an EO crystal and varies its polarization due to the Pockels effect. Measuring ellipsometrically these variations for different time delays between the pulses allows one to map the terahertz waveform. Recently, a non-collinear non-ellipsometric method of EO sampling has been proposed [1]. In this method, crossing of the probe and terahertz beams at the Cherenkov angle provides phase-matching and terahertz field is extracted from the intensity modulation of the probe beam without any polarization optics. The modulation results from angular separation of the probe beam parts, which are oppositely modulated by sum- and difference-frequency-generation processes [2]. The method was experimentally demonstrated by using a structure consisting of a $LiNbO_3$ crystal and a Si-prism-coupler [1]. Here, we implement non-collinear non-ellipsometric EO sampling in a GaAs crystal without any coupling optics. The results are compared with non-collinear ellipsometric EO sampling in GaAs [3].

We used the ellipsometric experimental setup described in [3]. The setup was modified by adding a non-ellipsometric arm, where a D-shape mirror divided the probe beam after the GaAs crystal into two halves and directed them to a balanced photodetector. Switching between the ellipsometric and non-ellipsometric arms was provided by a flipped mirror.

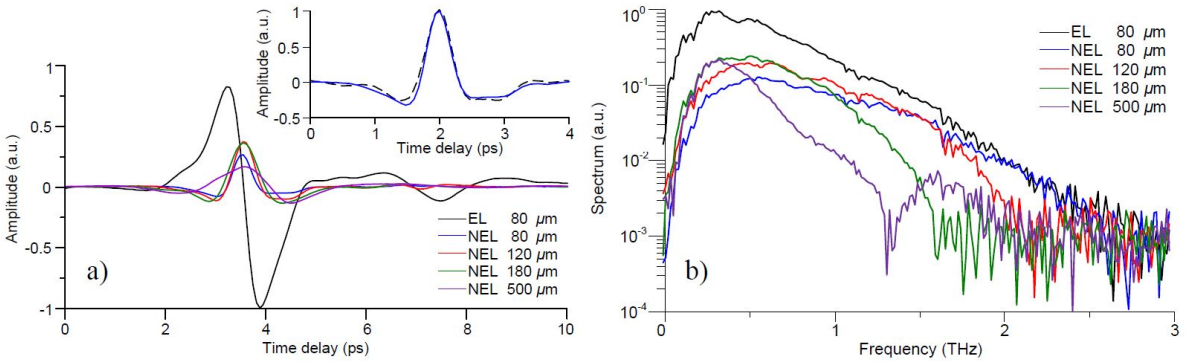

Figure 1: EO signals (a) and corresponding terahertz spectra (b) obtained by nonellipsometric (NEL) sampling with different probe beam widths. The spectrum obtained by ellipsometric (EL) sampling is shown for reference. Inset: normalized NEL 80 μm signal (solid) in comparison with the time derivative of the EL signal (dashed).

Fig. 1 shows the EO signals [Fig. 1(a)] and corresponding spectra [Fig. 1(b)] obtained by using the non-ellipsometric method for different widths (80, 120, 180, and 500 μm) of the probe beam. The detection efficiency is comparable to that of the ellipsometric method [Figs. 1(a) and 1(b)] but the signal shape is significantly different [Fig. 1(a)]. For the 80-μm wide beam, the shape is close to the time derivative of the ellipsometric signal [inset in Fig. 1(a)]. For the wider beams, the non-ellipsometric signal becomes smoother.

The results in Figs. 1(a) and 1(b) confirm the theoretical prediction [2] that the non-collinear non-ellipsometric scheme operates as a bandpass filter, whose transmission band is defined by the probe beam width.

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Three body attachment of electrons in humid air

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We report the results of theoretical and experimental study of the characteristic lifetime for three-body attachment of electrons in the low-density ($\sim 10^{12}$ cm^{-3}) plasma produced by 100 fs UV laser pulse in moist and dry air in the external DC electric field ranged from 0.2 to 8 kV/cm. In humid air characteristic electron lifetime is decreased twice compared to the dry one.

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Modeling femtosecond laser-induced electron dynamics in dielectrics by means of Optical Bloch Equations

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Modern laser technologies provide high-intensity single- or few-cycle laser pulses which open new doors for studies of laser-matter interaction processes. To predict new routes towards their active control, advanced theoretical and numerical models are required. When approaching the highly non-linear interaction regimes close to the material damage threshold, the traditional perturbation expansion of the polarization response is not valid anymore and a quantum-mechanical modeling is required [1-3]. A good candidate for this role is the Optical Bloch Equations (OBEs) approach, which provides all-order material response within a single self-consistent description. We develop a new OBEs-based model of laser matter-interaction which includes field-induced ionization, impact ionization and various relaxation processes taking place in dielectric materials. This model accounts for the linear as well as nonlinear polarization response leading to high harmonics and THz generation. Here the model is applied to describe the laser induced electron dynamics in various dielectric and wide-band gap semiconductor materials by intense femtosecond pulses.

In most of the current studies, Maxwell's equations or the unidirectional pulse propagation equation are coupled to the material response governing equations. The computation of the energy which is gained by the electron subsystem during the laser-matter interaction and the subsequent electron-lattice energy relaxation process are the keys for prediction of the laser induced damage threshold (LIDT) and controllable modification of material [5,6]. For the description of the laser-induced electron population in the conduction band, we employed the OBEs. The energy separation between the lowest and the highest considered conduction bands (CBs) is taken to be more or equal to 1.5 of the bandgap energy E_g [4], allowing to introduce the impact ionization term phenomenologically with a characteristic timescale τ_{imp}.

In this work we assume that the energy density required for material damage is: $\Delta Q = \frac{\rho}{M}\left(\int_{T_0}^{T_m} C(T)dT + L\right)$, where T is the material temperature, M is the molar mass and ρ is the density of the material, $C(T)$ is the material heat capacity per mole, T_0 is the room temperature and T_m is the material melting temperature. In case of crystalline materials, a latent heat L per mole is added. In fused silica the estimated energy density threshold for material melting is $\Delta Q = 2 - 4.32 kJ/cm^3$. In simulations, we calculate the energy density accumulated in the electron subsystem at the end of the laser pulse: $E = n_0 \sum \rho_e^i (E_i - E_1)$, where i goes from 1 to the considered number of CBs, E_i is the energy of the band with respect to the energy of the VB, n_0 is the neutral density. Here $i = 0$ corresponds to the VB, and i>0 corresponds to the CBs. The LIDT is reached if the energy density accumulated in the electron subsystem at the end of the laser pulse equals the estimated energy density required for the material damage: $E = \Delta Q$.

Let us consider the energy density gained by the electron subsystem of silica glass under an irradiation by a 10-cycles laser pulse with peak intensity from 10^9 Wcm^{-2} to

5×10^{14} Wcm^{-2} and photon energies from 0.5 eV up to 3 eV. First, the MRE approach [4] was used to calculate the electron density and corresponding energy density gained by the electronic subsystem at the end of the laser pulse (Fig.1 a). Second, the OBEs-based model with the same laser pulse parameters, material energy levels and τ_{imp} was used to calculate the energy density gained by the electronic subsystem at the end of the laser pulse (Fig.1 b). The energy density obtained with OBEs has pronounced minima at photon energies corresponding to the resonant multi-photon transitions from VB to the lowest CB. Particularly, for photon energy equal to 1.5 eV the LIDT is reached at the peak laser pulse intensity about 3.5×10^{14} Wcm^{-2} and corresponding fluence about 1.3 J/cm^2 (Fig.1 c).

The main result obtained by the developed approach is the description of the electron density and electron-subsystem energy evolution along the high-intensity femtosecond laser pulse in dielectric materials allowing to reproduce the LIDT. The obtained LIDT predictions are in good agreement with experimental observations [7,8].

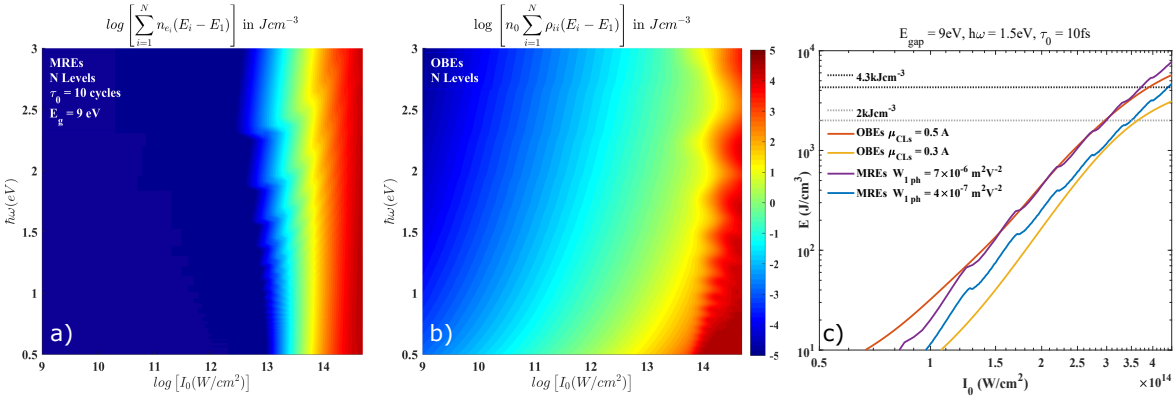

Figure 1: a) Energy density gained by the electron subsystem of silica glass under an irradiation by a 10-cycles laser pulse calculated by means of MREs. b) Energy density gained by the electron subsystem of silica glass under an irradiation by a 10-cycles laser pulse calculated by means of OBEs. c) Energy density gained by the electron subsystem of silica glass depending on the incident pulse peak intensity calculated for 10-fs 1.5-eV laser pulse both by means of MREs and OBEs. The impact of free parameters is illustrated. In OBEs the coherence relaxation time between the highest level and all other levels is set to 1 fs. Impact ionization is included with $\tau_{imp} = 1fs$ both in OBEs and in MREs. The bandgap of fused silica is taken 9eV.

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Quantum-mechanical elaboration for the description of low- and high-order harmonics generated in extended gas media

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The interaction of intense femtosecond laser pulses with the atomic and molecular gases or plasmas provides an effective source of bright coherent radiation in a wide range [1]. In the case of a femtosecond IR driving field this range extends from terahertz to X-Ray region including the generation of low and high harmonics (LHHG). The consistent solving of the LHHG description problem can only be obtained on the basis of the macroscopic theory, because the material properties of media change drastically the harmonic spectra with respect to the spectra emitted by a single atom. Therefore, a consistent theory of harmonic spectra should be based on a precise solution of the emitted radiation propagation problem and an exact solution of the single atom generation problem. However, both of these problems are very cumbersome and their analyses require a lot of computing time. Therefore, some approximate schemes which simultaneously take into account the single atom evolution dynamics and the medium dispersion properties are of great interest for practical applications.

Here, we propose the interference model based on the use of a 1D chain of atoms interacting with a laser field. The number of atoms depends on the pressure of the gas and the length of the gas cell. The response of each atom is calculated in strongly consistent way on the base of non - perturbative theory [2]. The integral field emitted by the atomic chain is the sum of single atom response fields. In spite of the relative simplicity of the model the calculated temporal and angular spectra can be directly compared with the experimentally registered ones [3].
We have investigated spatial and integral characteristics of the generated radiation with changing gas pressure and medium length. The dependence of the integral harmonics intensity on the gas pressure demonstrates peak structure. The results of numerical simulations have shown that there is a simple analytical formula connecting the following parameters: harmonic order (H), medium length (l) and gas pressure, at which the intensity of harmonic is maximum (p_{opt}):

$$l \cdot H \cdot p_{\text{opt}} = Const$$

The formula can be used for the optimization of the harmonic intensity in experiments. The model has been applied for the description of a rather complicated recent experiment presented in [4].

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Second and third optical harmonics generation in photonic crystals under femtosecond laser pulses excitation

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Optical harmonics and plasma emission were detected in globular silica photonic crystals when excited by 250 fs laser pulses at a power density up to 1 TW/cm^2. A Yb:KGW laser with a wavelength of 1026.0 nm was used as an exciting radiation source. At a pulse duration of 250 fs and a pulse repetition rate of 200 kHz, the peak radiation power of the fundamental harmonic at the femtosecond laser output was 0.7 GW. Laser emission was focused onto the photonic crystal surface to light spot of about 100 μm. As samples to be studied, synthetic opal matrices structured as a close-packed cubic lattice of silica balls (globules) 220, 240, 260, and 290 nm in diameter were chosen. Pores of opal matrices with a globule diameter of 240 nm were filled with ferroelectrics sodium nitrite and barium titanate. We note that sample degradation was not observed during irradiation of the photonic crystal surface for several hours. This is explained by the transparency of used samples in the visible spectral region and by the low energy of each laser pulse (5 μJ).

The third optical harmonic with a conversion efficiency of about 10% arose for the opal matrix containing pores filled with air. In globular photonic crystals filled with ferroelectrics, the second optical harmonic was observed with a few percent conversion efficiency. The highest conversion efficiency for exciting radiation of 1026 or 513 nm wavelengths was achieved when the frequencies of the exciting radiation or the optical harmonic were close to stop band edge.

The formation of enhanced generation of the third harmonic can be associated with the effect of a decrease in the group velocity of pump radiation propagation in the photonic crystal when operating near the band gap and with the effect of structural light focusing in the photonic crystal bulk. Structural light focusing in the globular photonic crystal leads to the formation of the regions with high intensity of optical radiation. Exactly when the pump wavelength coincides with the photonic crystal band gap position, high-intensity optical field regions are localized at the crystal globule center. In this case, the intensity of formed optical field caustics exceeds initial one outside crystal by several order of magnitude. Due to the formation of local high-intensity regions, high-efficiency medium pumping can be achieved.

Thus, the using of femtosecond laser pulses with high power density allows to observe very effective second and third optical harmonic generation in photonic crystals.

Quasistatic fields propagating ahead of ultrashort laser pulses in electro-optic crystals

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Optical rectification of ultrashort laser pulses in electro-optic crystals is a commonly used technique for generating pulsed terahertz radiation. Demand for stronger terahertz electric and magnetic fields for a variety of applications requires increasing pump intensity. At high pump intensities, multi-photon absorption becomes essential. It leads not only to the depletion of the pump but also to the generation of free carriers that absorb terahertz waves. Thus, free-carrier generation is commonly considered as a destructive effect for terahertz generation.

Recently, however, it was found that concurrent processes of optical rectification and free carrier generation can give rise to a positive effect, namely, the creation of quasistatic electromagnetic fields propagating ahead of the pump laser pulse, i.e., a quasistatic precursor [1]. The precursor is generated by a burst of an electric current, produced by the acceleration of the newly born carriers by the electric field of the rectified laser pulse. The magnitude of the precursor can exceed the field of the terahertz pulse generated behind the laser pulse. The consideration in Ref. [1] was restricted by the approximation of an infinitely wide undepleted laser beam. Here we investigate how a finite beam width and pump depletion affect the precursor generation. We also show that pumping by a chirped laser pulses allows one to avoid the negative effect of the pump depletion.

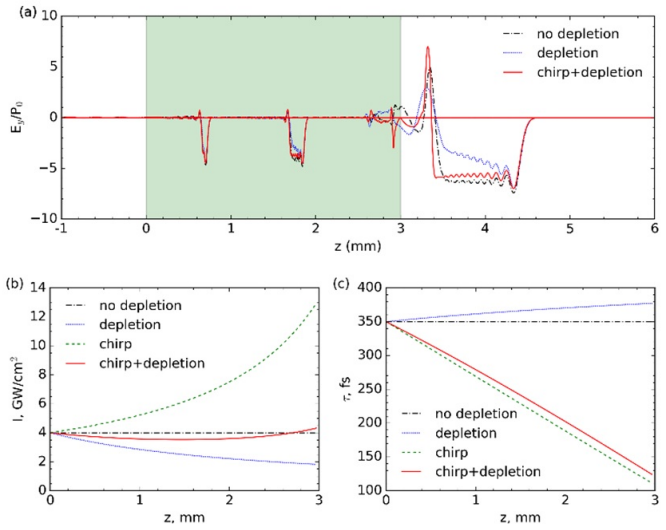

Figure 1: (a) Snapshots of E_y/P_0 at three moments of time for the cases when pump depletion is neglected, included, and compensated by chirping the laser pulse. The crystal (shaded region) is 3 mm thick. The arrows depict the position of the pump pulse. (b) Intensity and (c) 1/e-duration of the pump pulse as a function of the distance in the crystal.

Figure 1(a) shows snapshots of the electric field E_y generated by a Ti:sapphire laser pulse (780 nm wavelength) in a 3-mm thick GaP crystal. The field is normalized to the

amplitude of the nonlinear polarization P_0 induced by the laser pulse in the crystal. If pump depletion is neglected, the precursor has a form of a plateau, whose size increases with distance in the crystal and due to transmission through the crystal boundary. Pump depletion leads to an attenuation of the rear part of the precursor.

Due to dispersive compression of a negatively chirped laser pulse [Fig. 1(c)], the optical intensity can be sustained practically constant in the crystal [Fig. 1(b)]. This provides efficient precursor generation with almost the same shape as in non-depletion approximation [Fig. 1(a)].

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Molded filaments and applications

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The nonlinear propagation of ultrashort laser pulses in the form of solitons, filaments and light bullets is an exciting research field [1]. Beyond the basic studies on the complex spatio-temporal phenomena involved, the field is driven significantly by its numerous applications, like for example in materials engineering, remote spectroscopy [2], but also for their use as powerful secondary sources across the electromagnetic spectrum [3].

Here we discuss our recent advances in molding the shape, temporal and spectral properties of filaments [4] and some corresponding applications enabled through these advances. We demonstrate how it becomes possible, for the first time after 20 years of research, to achieve localized and controlled modification of the index of refraction in the bulk of silicon [5]. This advance opens the way for laser processing in the exciting field of silicon photonics.

We also discuss our recent advances in developing intense THz secondary sources using tailored laser filaments. We demonstrate that one may obtain powerful THz radiation using unconventional media, like liquids, where the medium presents strong linear absorption [6]. The mechanism responsible for this counterintuitive result is a phase locked second harmonic component in the filament that results in strong transient electron currents that radiate intense THz fields. Finally, we will also be discussing the way in achieving extreme THz electric and magnetic fields, in excess of GV/cm and kT strengths respectively, using intense two-color mid-infrared filaments [7].

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Control of multiple filamentation of TW IR radiation propagating along an air path by means of a deformable mirror

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The results of experimental studies on the nonlinear propagation of high-power femtosecond pulses of a Ti:Sa laser (carrier 780 nm, peak power ∼1 TW, energy ∼40 mJ) in a multiple filamentation mode along a 140-m air path are presented. At the path start, a multi-element reflector was mounted, consisted of a telescope and a flexible mirror (bimorph, 57 elements), which made it possible to form a radiation wavefront profile specified. The main task of the experiments was to study possibilities of controlling the position and structure of the filamentation domain of a wide-aperture laser beam (aperture diameter 5 cm) by introducing controllable aberrations of the wave phase profile.

Experiments have shown that, in comparison with conventional parabolic focusing, the use of a bimorph deformable mirror allows moving the filamentation and plasma formation domain throughout the path in a wider range of distances. In addition, it becomes possible to configure the spatial arrangement of high-intensity light channels (filaments) in a beam cross section. The plasma-free propagation mode of radiation in the form of an ordered bundle of high-intensity ($10^{11} - 10^{12}$ W/cm^2) weakly converging (< 0.01 mrad) long (> 100 m) light channels with millimeter diameters has been revealed for a number of special configurations of the beam wavefront.

The simulation of multiple filamentation of laser radiation with aberration phase front on the basis of numerical solution of the nonlinear Schrödinger equation in 3D configuration has shown that ordering of filaments in a beam occurs due to the appearance of stable (ring) light structures in it already during the self-focusing stage. The shape and energy reserve of these structures determine the arrangement and number of the forming intense channels, some of which then evolve into filaments accompanied by plasma channels.

Section 5: Femtosecond laser photobiology and photochemistry

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Program committee:
Andrei Razjivin (MSU, Russia)
Ivan Pozdnyakov (ICKC SB RAS, Russia)
Evegeny Ryabov (IS RAS, Russia)

Scope
Applications of Femtosecond spectroscopy in Chemistry and Physics
Ultrafast dynamics in molecules and clusters
Ultrafast phenomena in Biophotonics and Photobiology
Ultrafast spectroscopy of primary processes of photosynthesis

Ultrafast X-ray and optical studies of chemical and biological systems

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In this talk, I will present case studies of the photophysical and photochemical dynamics of coordination chemistry complexes and proteins using a toolbox of ultrafast methods: transient absorption from the infrared to the deep-ultraviolet, X-ray absorption and emission spectroscopy and photoelectron spectroscopy of liquid solutions. The processes to be discussed include: ferric impulsive electronic-to-vibrational energy conversion in ferric hexacyanide[1], photoaquation[2] in ferrous hexacyanide, transfer of vibrational coherence in ultrafast intersystem crossing events[3], and tryptophan to heme electron transfer in hemoproteins.[4]

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Deciphering the ultrafast dynamics in Zr-based MOFs

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Metal-Organic Frameworks (MOFs) are crystalline hybrid materials whose excellent physical and chemical properties have allowed their implementation in different fields of science and technology. Recently, the synthesis of new kind of photoactive MOFs have boosted their use in fields as photocatalysis or optoelectronics [1]. Some MOFs have shown that when the organic linkers are excited, a photoinduced ligand-to-cluster charge transfer (LCCT) is produced, leading to a charge separation state with electrons (e^-) and holes (h^+). These photogenerated e^- and h^+ can reduce or oxidize different chemical compounds as well as produce H_2 [2]. In the same line, the generation of these charge carriers is essential to further employ MOFs as electroluminescent layers in optoelectronic LED devices [3]. However, the photogeneration of LCCT in Zr-MOFs has been a subject of ongoing discussion.

Herein, armed with ultrafast pump-probe femtosecond fluorescence up-conversion and transient absorption techniques, we have unraveled the interactions that take place in the excited state of Zr-MOFs, as well as the photogeneration of a LCCT followed by a slow e^--h^+ recombination from the trap states of Zr metal clusters [4-5]. In this sense, we have investigated the photoprocesses taking place in the excited state of a high concentrated Zr-NDC (NDC = 2,6-naphthalene dicarboxylate) MOF suspension, where we showed intraparticle excimer-like interactions occurring in 3-5 ps [4].

In addition to this, we have also demonstrated spectroscopic and photodynamically the LCCT from NDC and NADC (amino-2,6-naphthalene dicarboxylate) linkers to Zr metal cluster parts in Zr-NDC and Zr-NADC MOFs [5]. The LCCT takes place in 60 fs for Zr-NDC MOF while it is in 170 and 60 fs for Zr-NADC in N,N-dimethylformamide (DMF) and dichloromethane (DCM) suspensions (Figure 1), respectively. To the best of our knowledge, this was the first time that the early dynamics of the LCCT process in Zr-MOF systems was established. Further evidences of the photogenerated (e^- and h^+) are given by the interaction of these MOFs with electron donor (N,N,N',N'-tetramethyl-p-phenylenediamine, TMPD) and scavenger (methyl viologen, MV2+) compounds. Our results shed light on the ultrafast processes of MOFs opening the way to better design these materials for photocatalysis and optoelectronic applications.

This work was supported by the MINECO through projects: MAT2014-57646-P and MAT2017-86532-R.

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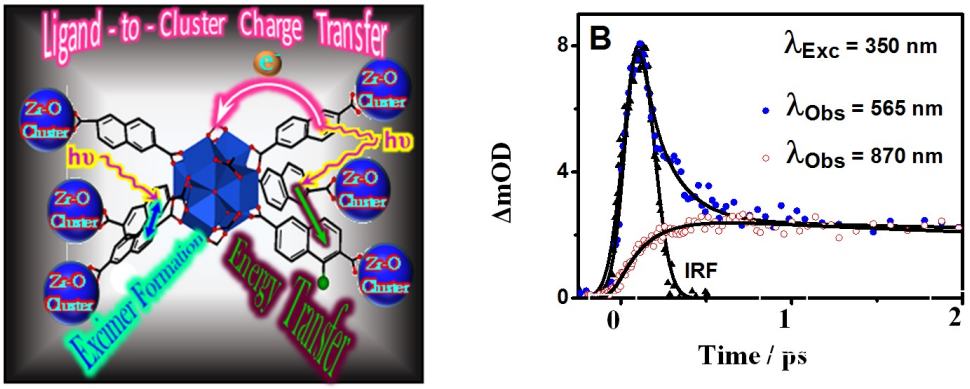

Figure 1: A) Schematic illustration of the processes that take place in photoexcited Zr-NADC MOF. B) Fs-transient absorption decays of Zr-NADC in DMF suspension exciting at 350 nm and observing as indicated.

One-dimensional optimization of the calculation of the probability of electron transitions taking into account one high-frequency vibrational mode

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A general formula is obtained for describing the probability distribution over the reaction coordinate and it is adapted to the reproduction of non-stationary fluorescence spectra. A program is developed that solves the optimization problem over one of the model parameters for the best fit of the experimental data. The spectra obtained using this model were compared with empirical data. Optimization was carried out for one of the relaxation times by Newton's method with fixed other parameters. In the process of optimization, the discrepancy between the theoretical and experimental curves was minimized for the corresponding instant of time. Automation of this kind of fitting, especially when expanding to a multivariate case before optimization by several parameters, is much more effective than fitting the graphs per oculum. The obtained law authentically reflects spectral dynamics, and also by means of methods of optimization of data growth with empirical data. This model is applicable for various polar solvents and soluble substances. The study was performed by a grant from the Russian Science Foundation (Grant № 16-13-10122).

Photophysics and photochemistry of platinum group metals complexes

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Combining methods of stationary and nanosecond laser flash photolysis and ultrafast kinetic spectroscopy one can reach complete understanding of mechanisms of photochemical reactions, starting from the absorption of a light quantum to the formation of final reaction products.

Realizing this approach we have studied photochemistry of several model hexahalide complexes of platinum metals with the common formula $M^{IV}Hal_6^{2-}$ in aqueous solutions. In all cases, photoaquation is the final photochemical processes. For $Pt^{IV}Br_6^{2-}$, $Ir^{IV}Cl_6^{2-}$ and $Os^{IV}Cl_6^{2-}$ it occurs via heterolytic M-Hal bond cleavage, completing within time region of tens of picoseconds. In the case of $Pt^{IV}Cl_6^{2-}$ the primary photochemical process is an inner-sphere electron transfer. The occurring long-living Pt(III) intermediates are involved to the chain aquation process. For the case of $Os^{IV}Cl_6^{2-}$ the combination of ultrafast pump-probe spectroscopy and quantum chemical calculations of electronic absorption spectra of possible intermediates allowed us to build the completely proved reaction mechanism.

The second group of systems studied in this work were diazido Pt(IV) complexes, *cis*, *trans*, *cis*-$[Pt(N_3)_2(OH)_2(NH_3)_2]$ (1) and *trans*,*trans*,*trans*-$[Pt(N_3)_2(OH)_2(NH_3)_2]$ (2), and complexes of Ru(II), *cis*, *fac*-$[RuCl_2(DMSO)_3(H_2O)]$ (3) and *trans*, *cis*, *cis*-$[RuCl_2(DMSO)_2(H_2O)_2]$ (4). All these complexes were tested as light-activated agents in photodynamic therapy (PDT) of tumors. Mixed-ligand Pt(IV) and Ru(II) complexes are considered to be prodrugs providing cytotoxic species under the action of light. The final products can bind DNA like well-known anti-cancer drug cisplatin. In spite of practical importance, information on the mechanisms of photochemical processes for mixed-ligand complexes of platinum metals in the literature is scarce

The process of photolysis of (1) and (2) is multistage. The first stage is photosubstitution of an azide ligand to a water molecule. This process was shown to be a chain reaction involving redox stages. The primary photochemical process is an electron transfer from an azide ligand to the central cation followed by a release of an azide radical to the solution bulk. Several Pt(IV) and Pt(III) intermediates responsible for chain propagation were registered. The mechanism of photoaquation is proposed. Prolonged photolysis results in photoreduction of Pt(IV) to Pt(II). The light-induced cytotoxicity of the complexes is probably the combination of two factors, namely (i) a cisplatine-like effect of Pt(II) products and (ii) an azide radical reactivity.

Irradiation of complex (3) results in the reaction of photaquation / rearrangement to complex (4). The total reaction time is about 30 ps. Based on the data of ultrafast pump-probe measurements, the mechanism of photolysis is proposed. Prolonged irradiation of (3) and (4) results in further aquation followed by thermal oxidation by dissolved oxygen. Aquated species of Ru(II) can bind to DNA providing anti-tumor effect similar to that of cisplatin.

The work was supported by the Russian Scientific Foundation (grant № 18-13-00246).

Photoinduced electron transfer via exciplex formation in diazaporphyrin-porphyrin and porphyrin-pyrene dyads

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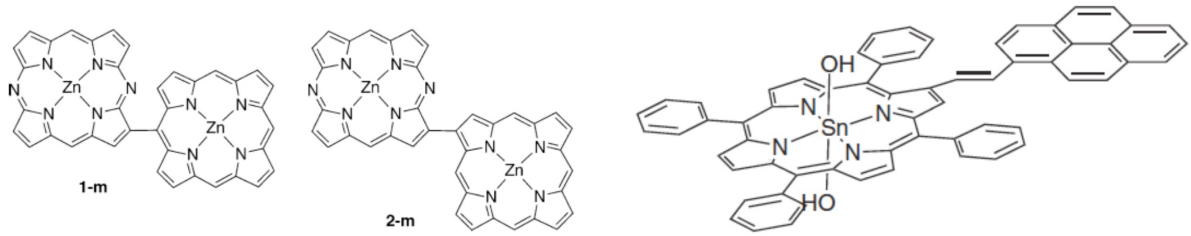
Two hetero dimers of diazaporphyrin-porphyrin (DAP-P, in $\beta-$meso and $\beta-\beta$ configurations) and Sn(IV)tetraphenylporphyrin-pyrene (T-P, connected by a β-vinyl linker) and their corresponding precursors, were studied in solutions of different solvent polarities using steady-state and time-resolved absorption and emission measurements at ps and fs time-scales.



$\beta-$meso (I-m) and $\beta-\beta$ (2-m) conformers of DAP-P Tetraphenylporphyrin-pyrene of T-P

For both DAP-P dimers long-living (1.4 ns) exciplexes were formed in non-polar toluene, but they both were preceded by short-living (0.6 -17 ps) fluorescent primary exciplexes. The long-living exciplex formed a charge-transfer dyad, with a lifetime more than 5 ns. In a more polar benzonitrile the primary exiplexes (2.4 ps and 12 ps) were formed, as well, but the shorter-living relaxed directly to the charge transfer complex, with a life-time (180 ps), shorter than that of the exciplex (660 ps), formed from the longer-living primary exciplex.[1]

When the pyrene moiety of the T-P dimer was excited, an efficient excitation energy transfer from pyrene (P) to porphyrin (T) took place leading to a quenching of the pyrene emission and to the enhanced porphyrin emission. A new emissive excited state complex (T-P)* was observed irrespective of the chromophore being excited. The emission was more pronounced in non-polar hexane showing a mono-exponential decay of the porphyrin emission. When excited the porphyrin moiety at $\lambda_{ex} = 425$ nm, the fs transient absorption analysis showed two different intermediate species ($\sim$7-11 ps and 80-100 ps) with broad absorption in the near-IR region. This implied either the existence of two different excited conformers ^{1}(a-T-P)*, and ^{1}(b-T-P)*, which decayed to the ground state via a charge separated state, or the formation of the (T-P)* state via the second excited state of the porphyrin moiety ^{2}(T*-P)*, yielding first a vibrationally excited emissive ^{v}(T$-$P)* state, with a lifetime of 80-100 ps [2].

The mechanisms of the electron transfer is discussed in in frame of the exciplex forming.

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Exited-state dynamics of $[CuCl_4]^{2-}$ and $[CuBr_4]^{2-}$ complexes in solution

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We have presented detailed description of the ultrafast dynamics of $[CuCl_4]^{2-}$ and $[CuBr_4]^{2-}$ complexes in acetonitrile upon excitation into all possible d-d/ligand field (LF) electronic excited states upon NIR excitation, and into electronic excited ligand-to-metal charge transfer (LMCT) states upon UV-vis excitation. LF states are found to be nonreactive states, while LMCT excitation leads to the ionic dissociation of studied complexes. We revealed that the behavior of electronically excited $[CuBr_4]^{2-}$ complex is almost identical to $[CuCl_4]^{2-}$ complex in acetonitrile with some difference in rate constant values.

We have demonstrated that $^2A_1$, $^2B_1$, and ^2E LF excited states directly relaxes into the ground state, their lifetimes are equal to 5, 1, and < 0.15 ps for $[CuCl_4]^{2-}$, and 6, 1.5, and < 0.3 ps for $[CuBr_4]^{2-}$ respectively. Addition of water to acetonitrile solution of $[CuCl_4]^{2-}$quenches the $^2A_1$ LF excited state due to the efficient electronic-to vibrational energy conversion resulting in excitation of O-H stretching modes of the extracoordinated H_2O molecule. We found that the nonradioactive rate constants for the transitions between LF states obey the energy gap law for weakly coupled potential energy surfaces and decrease with increase the energy gap between LF states. Meanwhile, extremely fast internal conversion between ^2E LF excited state and $^2B_2$ ground state suggests the conical intersections between these electronic states. Indeed, the analysis of LF excited states PES using TDDFT methodology have demonstrated the presence conical intersection between $^2B_2$ ground state and ^2E LF excited state, whereas $^2B_1$ and $^2A_1$ LF excited states do not cross ground electronic state. The Franck-Condon mode is found to be symmetric "umbrella" X-Cu-X bending vibration.

In our experiments, $[CuCl_4]^{2-}$ and $[CuBr_4]^{2-}$ complexes were also excited LMCT states using 420 and 550 nm pump wavelengths, respectively. After formation, on a timescale less than 200 fs, LMCT state undergo internal conversion to ground electronic states though the lower-lying vibrationally hot LF excited states and Cu-X ionic bond dissociation, forming $[CuX_3]^{2-}$ and X^- ions. $[CuX_3]^-$ complexes are solvated by acetonitrile molecule to form $[CuX_3CH_3CN]^-$ complexes within first 20 ps, which than recombines into parent $[CuX_4]^{2-}$ complexes as a result of reaction with halide-ions by associative interchange mechanism. Bimolecular rate constants of the recombination process are the same for chloride and bromide complexes and are equal to $9 \cdot 10^7$ M^{-1} s^{-1}.

Calculations have been made with the assistance of the Saint-Petersburg State University Computer Center. Optical measurements were performed at the Center for optical and laser materials research of Saint-Petersburg State University.

Dynamic solvent effect and ultrafast charge recombination in excited donor-acceptor complexes

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Control of charge transfer processes requires knowledge of its detailed mechanism. Due to the large number of known mechanisms, the identification of the mechanism in specific systems is a challenge so far. The idea of how to distinguish between thermal and nonequilibrium modes of charge recombination in excited donor-acceptor complexes is proposed in this article. Simulation of ultrafast charge recombination kinetics in excited donor-acceptor complexes within the framework of the multichannel stochastic model has shown that a number of regularities inherent in nonequilibrium charge transfer strongly differs from the kinetic regularities of charge transfer occurring in the thermal regime. Among them there are opposite regularities, for example, the dependence of the dynamic solvent effect on the free energy gap. A method for identifying the charge recombination mechanism verification (thermal or nonequilibrium) in photoexcited donor-acceptor complexes is proposed. The study was performed by a grant from the Russian Science Foundation (Grant № 16-13-10122).

The effect of solvent relaxation time constants on free energy gap law for ultrafast charge recombination following photoinduced charge separation

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The regularities inherent in the kinetics of ultrafast charge recombination ensuing the photoinduced charge separation in donor–acceptor dyads in solutions are discussed in the report. The simulations of the kinetics have been performed within the well-known verified stochastic multichannel point-transition model. Increasing the solvent relaxation time scales has been shown to strongly vary the dependence of the charge recombination rate constant on the free energy gap. In slow relaxing solvents the non-equilibrium charge recombination occurring in parallel with solvent relaxation is very effective so that the charge recombination terminates at the non-equilibrium stage. This results in a crucial difference between the free energy gap laws for the ultrafast charge recombination and the thermal charge transfer. For the thermal reactions the well-known Marcus bell-shaped dependence of the rate constant on the free energy gap is realized while for the ultrafast charge recombination only a descending branch is predicted in the whole area of the free energy gap exceeding 0.2 eV. From the available experimental data on the population kinetics of the second and first excited states for a series of Zn–porphyrin–imide dyads in toluene and tetrahydrofuran solutions, an effective rate constant of the charge recombination into the first excited state has been calculated. The obtained very high rate constant is nearly invariable in the region of the charge recombination free energy gap from 0.2 to 0.6 eV that supports the theoretical prediction. The study was performed by a grant from the Russian Science Foundation (Grant № 16-13-10122).

Ultrafast spectroscopy of optical phonons and excitons in quantum dots

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The goal of the present talk is to report the coherent phonons dynamics in strongly confined quantum dots under varied the pump fluences and in the presence of redox quenchers of excitons. Main characteristics of coherent phonons (amplitude, frequency, phase, spectrogram) of QDs are reported. We for the first time demonstrate that the amplitude of coherent optical LO phonon at 6.16 THz excited in CdSe nanoparticles by femtosecond unchirped pulse shows non-monotone dependence on the pump fluence. This dependence exhibits the maximum at pump fluence $\sim$0.8 mJ/cm^2. Time frequency representation of oscillating signal corresponding to LO phonons revealed by continuous wavelet transform (CWT) shows the robust destructive quantum interference at the origin of sharp (optical phonon) and continuum-like (exciton) quasiparticles. CWT spectrogram demonstrates nonlinear chirp at short time delays, where the chirp sign depends on the pump pulse fluence. CWT spectrogram revealed the anharmonic coupling between optical and acoustic phonons. The fast quenching of the exciton by methyl viologen (MV^{2+}) on the surface of the quantum dot CdSe leads to the inverse piezoelectric effect, which causes an increase in the amplitude of the coherent acoustic phonon in the CdSe / MV^{2+} system compared to pure CdSe.

Work was supported by RSF 17-13-01506.

Energy transfer and trapping in photosystem I from Arthrospira platensis

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The pump-probe spectroscopy of the isolated photosystem I (PSI) from *Arthrospira platensis* have shown that the energy transfer from antenna to the reaction center at room temperature occurs within the scope of 40 ps. By modelling of the PSI kinetics and the linear spectroscopy, the exciton models of the PSI monomer and the trimer have been developed with the help of a multiparametric optimizer, which is called the Differential evolution.

Temperature dependence of the water oh-stretch band in the off-resonant raman spectroscopy: a computational approach

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Both picosecond and nanosecond off-resonance Raman spectroscopy measurements of the bulk water OH-stretch band (3200-3600 cm^{-1}) have revealed the same linear temperature dependence of the band centroid with the coefficient about 0.87 $cm^{-1}C^{-1}$. By applying the multimode Brownian oscillator model and modelling the Raman OH-stretch band temperature dynamics; we have calculated the characteristic spectral density of the intermolecular and intramolecular degrees of freedom in the bulk water.

Ultrafast dynamics induced by femtosecond laser radiation in mixed molecular clusters

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The results of the studies on dissociation dynamics of $(CF_3I)_n(CF_3Br)_m$ mixed molecular clusters induced by femtosecond UV radiation ($\lambda = 266$ nm) are presented. The time-resolved method of two-photon UV excitation and photoionizing probing ($\lambda = 400$ nm) combined with the time-of-flight (TOF) mass-spectrometry is applied.

$(CF_3I)_n(CF_3Br)_m$ mixed molecular clusters were formed during supersonic expansion through the pulsed nozzle. The mixture of CF_3I and CF_3Br gases was used with Ar carrier gas at room temperature and total pressure of 200 kPa above the nozzle. The clusters and the products of their UV induced decay were ionized by femtosecond laser radiation ($\lambda = 400$ nm) and thus formed ion products were then detected by means of TOF mass-spectrometer.

It is found that mixed clusters excited to Rydberg states lying below the ionization threshold are decomposed as a result of relaxation processes, giving rise to free neutral molecules of CF_3I and CF_3Br. Thus formed molecules are the origin of ion signal from CF_3I^+ and CF_3Br^+ molecular ions which grows on 100 ps time scale (see fig.1(a)). Simultaneously the signal from IBr^+ ions decays on the same time scale as the origin of this ion signal is $(CF_3I)_n(CF_3Br)_m$ mixed cluster (see fig.1(b)). As could be seen on Fig.1(b), experimental curve could be fitted by single exponential decay function with time constant of 1 ns, but this time constant could be used only for rather rough estimate on characteristic rate of relaxation processes.

Proposed mechanism of electronic–vibrational relaxation process includes several stages. Initial laser excitation of Rydberg states in clusterized molecules relaxes on 10 ps time scale with formation of lower lying excited electronic states of the molecules in the cluster, which are most likely vibrationaly excited. Further relaxation leads to migration of the vibrational energy stored in intramolecular vibrational modes to the bath of low frequency intracluster modes and subsequent growth of the cluster temperature. Finally, the cluster dissociates with formation of free neutral molecules.

This work was supported by the Russian Foundation for Basic Research, grant № 16-02-00139.

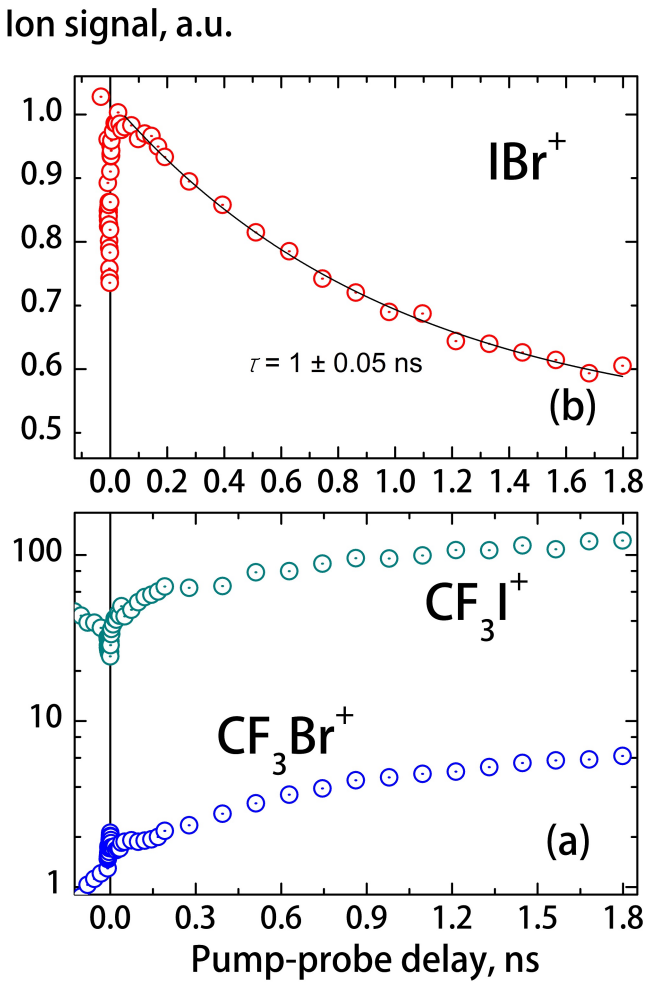

Figure 1: (a) Kinetics of CF_3I^+ and CF_3Br^+ ion signals from molecules formed as a result of the cluster decay; (b) kinetical curve of IBr^+ ion signal which indicates the decay of mixed clusters.

Ultrafast processes in photophysics of natural fulvic acids

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Photochemical processes playing important role in global cycles of organic nutrients in environment. Humic substances (HS, including humic and fulvic acids) are naturally photoactive components, which widely present in surface waters. These compounds are able to generate reactive oxygen species (ROS) under solar irradiation, which can react with dissolved organic pollutants initiating their degradation and mineralization. However initial stages of photophysics and photochemistry of HS which are responsible for further generation of ROS are still poor investigated. This talk presents results obtained by means of femtosecond kinetic spectroscopy with picosecond time resolution. Spectral and kinetic characteristics of HS excited singlet and triplet state have been obtained for the first time. Relation of these properties with efficiency of ROS generation is discussed.

The work was financially supported by Russian Foundation for Basic Research (grants № 17-03-00252, 18-53-53006_GFEN).

Molecular UV-filters of the human eye lens: the diversity of the ultrafast mechanisms of the excited state deactivation

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The human eye is protected from Solar UV light due to the presence of low molecular weight compounds in the eye lens – kynurenine (KN) and its derivatives. These intrinsic chromophores of the eye lens exhibit highly efficient deactivation of excited states with ultralow yields of reactive species like triplet states and radicals. Since the discovery of these molecular UV filters in the eye lens in the beginning of 70s (XX^{th} century), the mechanisms of effective deactivation of their excited states were unknown up to the recent time. This mystery was unveiled in our works. Fluorescence and transient absorption experiments with femtosecond time resolution have shown a variety of deactivation mechanisms in kynurenines: from excited state decay via inter-/intra-molecular hydrogen interactions to keto-enol tautomerisation and finally to conical intersection between potential energy surfaces of excited and ground states.

This work was supported by Grant Council of President of RF (MK-1515.2017.3) and RFBR project № 17-03-00656.

Primary processes in photophysics and photochemistry of diazide Pt(IV) complexes prospective for anti-cancer photodynamic therapy

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Pt^{IV} complexes are tested as prodrugs for anticancer photodynamic therapy, which can be provided in the absence of dissolved oxygen. Mechanism of action is based on photoreduction of Pt^{IV} to Pt^{II} complexes inhibiting DNA transcription and replication similar to cisplatin. The light-induced cytotoxicity of diazide Pt^{IV} complexes (1) and (2) (Fig. 1) is close to tcytotoxicity of cisplatin in dark [1].

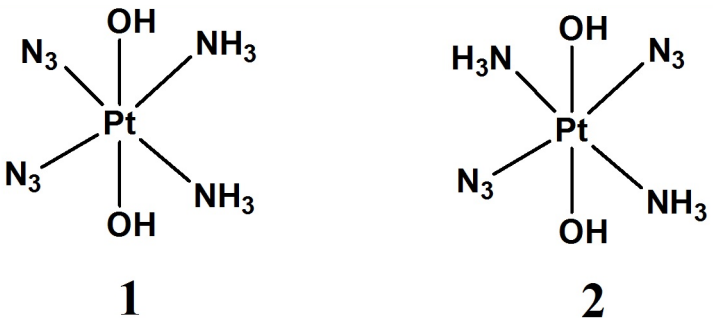

Figure 1: *cis*,*trans*,*cis*-$[Pt^{IV}(N_3)_2(OH)_2(NH_3)_2]$ (1) and *trans*,*trans*,*trans*-$[Pt^{IV}(N_3)_2(OH)_2(NH_3)_2]$ (2).

In this work primary photochemical processes were studied for aqueous solutions of (1) and (2) by means of steady-state photolysis, nanosecond laser flash photolysis and ultrafast kinetic spectroscopy (time resolution $\sim$ 100 fs). The process is shown to be multistage. The first stage is the photosubstitution of an azide ligand to a water molecule. This process was shown to be a chain reaction with the concentration-dependent quantum yield.

In the laser flash photolysis experiments the formation of two sequentially occurring Pt^{III} intermediates were recorded. Disproportionation of Pt^{III} intermediates leads to the formation of final Pt^{II} products, providing the therapeutic effect of the initial compounds. The reaction mechanism represented by equations (1-7) is proposed.

Chain initiation

$$[Pt^{IV}(NH_3)_2(N_3)_2(OH)_2] \xrightarrow{h\nu} [Pt^{III}(NH_3)_2(N_3)(OH)_2] + N_3^{\bullet} \quad (1)$$

$$[Pt^{III}(NH_3)_2(N_3)(OH)_2] \longrightarrow [Pt^{III}(NH_3)_2(OH)_2]^+ + N_3^- \quad (2)$$

Chain propagation

$$[Pt^{III}(NH_3)_2(OH)_2]^+ + [Pt^{IV}(NH_3)_2(N_3)_2(OH)_2] \xrightarrow{H_2O}$$
$$[Pt^{III}(NH_3)_2(N_3)(OH)_2] + Pt^{IV}(NH_3)_2(N_3)(OH)_2(H_2O)]^+ \quad (3)$$

Chain termination

$$Pt^{III} + Pt^{III} \rightarrow Pt^{IV} + Pt^{II} \quad (4)$$

$$Pt^{III} + N_3^{\bullet} \rightarrow Pt^{IV} + N_3^{-} \quad (5)$$

$$Pt^{IV} + N_3^{\bullet} \rightarrow Pt^{V} + N_3^{-} \quad (6)$$

$$Pt^{V} + Pt^{III} \rightarrow Pt^{IV} + Pt^{IV} \quad (7)$$

In the experiments on ultrafast pump-probe spectroscopy we observed the formation of intermediate absorption, most likely belonging to the lower electronic excited state (LE) of the original complex. The LE state is transformed to another intermediate interpreted as the radical complex (RC) formed by Pt^{III} species and an azide radical; the characteristic time of the RC formation is about 20 ps. In the time interval 0.1- 10 ps the RC is transformed to the Pt^{III} intermediate observed in the laser flash photolysis experiments. Therefore, the photosubstitution of N_3^- to a water molecule is the complicate chain process including at least five different Pt^{IV} and Pt^{III} intermediates.

Finally, photochemistry of complexes (1) and (2) was studied in time scale 100 fs -– 100 ms, i.e. from the absorption of a light quantum to the formation of final reaction products [2].

The work was supported by Russian Foundation of Basic Research (grant № 18-33-00009).

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Photoinduced charge separation at organic-semiconductor interface

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Photoinduced electronic interactions at organic-semiconductor interface has been a subject of intensive study for a few decade already, but the research has accelerated recently due to developments of nanostructures, multifunctional compounds, and hybrid solar cells. The ultrafast spectroscopy techniques, and in particular femtosecond pump-probe methods, are experimental basis to study and understand the photophysics of interactions and carrier dynamics at the interface. The presentation will focus on two semiconductor nanostructure, ZnO nanorods [1] and TiO_2 nanoparticles [2], functionalized by porphyrin and phthalocyanine derivatives using self-assembled monolayer (SAM) method. This selection is inspired by the most widely used compounds and structure in dye sensitized and hybrid solar cells. The organic dye sensitizer absorbs light and injects electron to the semiconductor in subpicosecond-picosecond time domain. This process depends strongly on the linker and aggregation tendency of the dye sensitizer [3-4]. However, we found that for one and the same dye the electron injection is faster to TiO_2 than to ZnO, but the electron-hole pairs recombine is faster with TiO_2 nanopartcles.

The recombination rate at the organic semiconductor interface can be strongly suppressed by adding a hole transporting material (HTM) on top or the sensitizing layer. HTM picks the holes from the sensitizer and deliver them to an external electric circuit in complete solar cell device. We have tested two HTMs, polythiophene derivative (P3HT) and a small molecule known as Spiro-OMeTAD (Spiro), and shonw that the HTM may change the photophysics at the interface quite drastically [4-5]. The P3HT layer picks hole from the sensitizer with average time constant of few tens of picosecond depending on the dye. On the contrary, the Spiro layer comes in closer contact with the sensitizer and switches the primary charge separation to dye-HTM interface, which is completed in a fraction of picosecond and leaves an electron in the sensitizer (creating dye anion) and a hole in HTM. This is followed by electron injection from the anion to the semiconductor with roughly 100 ps time constant, thus resulting in the same long distance charge separation with electrons in the conduction band of the semiconductor and hole in the bulk of HTM.

The ultrafast spectroscopy is indispensable tool in study and optimization of the primary photo-reactions at organic-semiconductor interface. It brings up the key photophysics knowledge and provides hint for the device optimization.

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Ultrafast photoinduced symmetry-breaking charge transfer

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A substantial number of molecular systems are characterized by two, or more, energetically-equivalent photoinduced charge-transfer pathways. Upon photoexcitation, charge transfer occurs along one of these pathways, resulting in a breaking of the symmetry [1]. Over the past years, we have been investigating the dynamics of such processes to understand how symmetry is broken. Two different types of symmetry-breaking (SB) charge-transfer processes will be discussed:

1) Photoinduced SB charge separation between two identical molecules. Using ultrafast transient dichroism, we could show that the direction of the charge separation, i.e. hole or electron transfer, is determined by the environment [2]. These processes can be used advantageously for photovoltaic applications [3].

2) Excited-state SB in quadrupolar D-π-A-π-D or A-π-D-π-A molecules. Although symmetric, these molecules behave like strongly dipolar single-branch A-π-D dyads once in the excited state. The dynamics of this SB could be directly visualized using time-resolved IR spectroscopy. Our data revealed that SB is due to the environment and not to structural fluctuations [4-6].

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Section 6: Physics and technology of ultrashort laser pulses and innovative femtosecond laser technology.

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Program committee
Alexander Konyashchenko (LPI, Russia)
Nikita Didenko (LPI, Russia)

Scope
Solid-state, parametric, and hybrid laser systems
Stretchers, compressors, and phase control
Diagnostic and measurement techniques
Innovative femtosecond laser technology

Several technological approaches for new generation of ultra-high peak and average power Ti:Sapphire laser systems

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Laser systems with up to 100 kW average power and a peak power of 100s TW regime are require for successful applications of laser–plasma particle accelerators, high harmonic generation, high speed and precision micro/ nano-machining in industry and medicine [1]. There are several laser technologies capable to achieve these parameters, such as fiber laser, optical parametrical oscillators and amplifiers, thin disc (TD) lasers and others. This report will show that extraction during pumping thin disc (EDP-TD) Ti:Sa laser amplifiers are a promising candidates for reaching this goals. The results of design, development and proof-of-principal experiments with two types of EDPTD Ti:Sa amplifiers with reflection and transmission based optical schemes will be presented [2,3], as well as the simulations of TD amplifier modules for large peak power with double channel cooling. It was shown that increasing the disk diameter whilst maintaining the same aspect ratio leads to extremely efficient heat extraction and a flat temperature profile at a high repetition rate operation. A double disk configuration with three coolant channels was also modelled and showed that 40 J pumped amplifiers running at 100 Hz repetition rate could operate with peak temperature below 37°C with an average power of 2 kW.

Nevertheless, attempts to reduce the pulse energy and increase repetition rate while maintaining same average power have led to significant temperature rises due to reducing of the total volume of the gain media and so the ability of heating scattering. A reduction in the crystal size is needed in order to keep the seed fluence near the saturation one, which will eventually cancel the advantage the thin disk geometry. This talk will provide a solution to this problem exploiting the TD-slab geometry of the gain media [4], which also enables higher repetition rate and increased amplifier efficiency when direct pumped by CW laser diodes [5].

The high output peak power means also a short pulse duration, and so large spectral bandwides. As with any laser medium the bandwidth of Ti:Sapphire lasers is limited by gain narrowing. The modern developments that, applied to Ti:Sapphire medium, will allow to overcome the currently existing technological limits of bandwidth. Polarization encoded Chirped Pulse Amplification (PE-CPA) will be considered as one of the efficient method for spectral enlargement [6,7]. In the report the experimental results as well as a numerical simulations will be presented. The computer modeling of a PE-CPA Ti:Sapphire amplifier with seed pulses broader than the experimental bandwidth shows that a 200 nm spectrum is achievable at a multi-joule level. The combination of presented methods is able to support laser pulses of few oscillations at hundreds of TW peak and kW average power.

The electrical polarization of a laser crystal by an external electrical field can give a new possibilities for wide control of the laser parameters, increasing their range for existing lasers and create a new line of the laser devices. Previously, the direct impact of external electrical fields on the laser solid state gain media was estimated using as an example Ti:Sapphire crystal. It was demonstrated that metal-ligands distances displacement in this crystal under external electrical field can be comparable to that caused by mechanical pressure of 150 kbar, measured in early experiments. In this talk, the experimental results of the electrical field impact on Ti: Sapphire crystal optical parameters will be reported. These experiments demonstrate a reduction in the luminescence spectral

amplitude and an increase in bandwidth with the red-shift of the maximal amplitude after high voltage application.

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Flux density and spatio-temporal couplings in a collapsing electromagnetic wave

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The progress of femto- and atto-physics requires development of accurate measurement of pulse temporal characteristics. However, they vary significantly for ultrashort pulses in space from point to point [1,2] especially near focal region. Therefore, the methods for spatio-temporal characterization of ultrashort laser beams have been suggested and realized for lasers from regular [3-8] to PW scale power [9]. Among them are the methods of spatially resolved Fourier-transform spectroscopy [7,8]. The information acquired at such measurements can be used to control laser beam, for instance to increase the field intensity at the focal area [9]. On the other hand, not arbitrary spatio-temporal field configurations can be found in Nature, but only those satisfying Maxwell equations, and they are preferable to be used for spatio-temporal methods of measurements and beam characterization. In this paper we investigate an exact analytical solution of electromagnetic field equations in the free space and corresponding spectrum and flux density in the near and far zones. Maxwell field equations in free space can be reduced to two equations for vector potential $\vec{A}$:

$$\Delta\vec{A} - \frac{1}{c^2}\frac{\partial^2\vec{A}}{\partial t^2} = 0,\ div\vec{A} = 0 \quad \text{with } \vec{E} = -\frac{1}{c}\frac{\partial\vec{A}}{\partial t},\ \vec{H} = rot\vec{A} \tag{1}$$

An evident solution to (1) is:

$$\vec{A} = \vec{l} \times grad\ u, \tag{2}$$

where $u(\vec{r},t)$ similarly to $\vec{A}$ satisfies scalar wave equation (1) and $\vec{l}$ is a unit vector constant in space and time. Spherically symmetrical $u(\vec{r},t)$ can be written for arbitrary shape function f(x) as [10,11]:

$$u(r,t) = \frac{f(ct+r) - f(ct-r)}{r}. \tag{3}$$

Using (1)-–(3) one obtains $\vec{E}$ and $\vec{H}$ as exact solutions of Maxwell equations in the closed form:

$$\vec{E} = -\frac{1}{c}\frac{\partial\vec{A}}{\partial t} = -\frac{1}{c}\frac{\partial^2 u}{\partial t\partial r}\vec{l}\times\vec{n},\ \vec{H} = rot\vec{A} = (\Delta u - \frac{1}{r}\frac{\partial u}{\partial r})\vec{l} - \left[r\frac{\partial}{\partial r}\frac{1}{r}\frac{\partial u}{\partial r}\right](\vec{l}\vec{n})\vec{n},$$

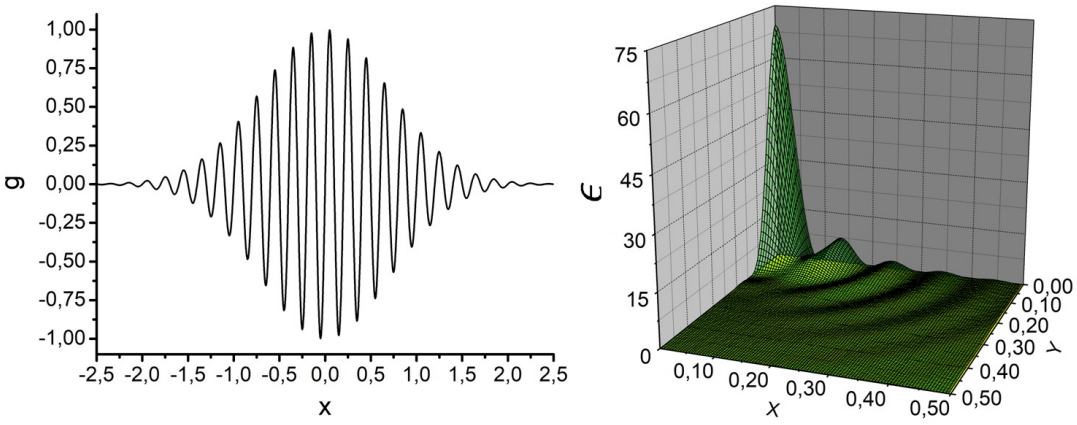
$$\Delta u = \frac{1}{r^2}\frac{\partial}{\partial r}(r^2\frac{\partial u}{\partial r}),\ \vec{n} = \frac{\vec{r}}{r} \tag{4}$$

If $f(x)$ vanishes at large $|x|$ the beam has finite energy. The light pulse (4) describes a shell arriving from large r, cumulating at r=0 and going back to $r \to \infty$. Formulas (3),(4) allow to obtain full spatio-temporal characteristics of the considered collapsing light beam. Spatially resolved spectrum and field behavior in time are found in [12]. These findings led to the following scaling of the energy density ϵ_m in the center of a collapsing pulse beam:

$$\epsilon_m = \frac{8\pi\sqrt{2\pi}}{3}\frac{\varepsilon}{(\lambda_0^3)}\frac{\delta\omega}{\omega_0},\ \lambda_0 = \frac{2\pi c}{\omega_0} \tag{5}$$

where ε is the total pulse energy, ω_0 and $\delta\omega$ are the central pulse frequency and linewidth.

Despite the obvious difficulties in practical implementing the high-symmetric space-time structure of the field (3), (4), a systematic comparison of the scaling (5) with the available experimental data seems to be of interest. Figure 1A demonstrates the shape of the field of a collapsing pulse far from the center (at $t \to \pm\infty$). Figure 1B shows 2D distribution of the energy density at the moment of maximum pulse field concentration ($t = 0$).



1A 1B

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Time and spectral pulse dynamics of a 2-μm hybrid laser during the amplification by holmium-doped fiber amplifier

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In the last decade, ultrafast fiber lasers operating in the spectral range of 2-3 μm have attracted attention due to a number of their possible technological and scientific applications. For example in wireless communications, supercontinuum generation, gas sensing, material processing, healthcare and so on [1]. However, for some industrial or technological applications, the energy of the pulses of such lasers is not enough. Therefore, the goal of ultrashort pulses amplifying has arisen.

Thus, in this work, we demonstrate the time and spectral dynamics of 2–μm ultrashort pulses changes depending on the value of amplification and intracavity dispersion.

The ring cavity of the master oscillator (MO) was formed by Ho-doped active fiber and single-mode fiber (SMF) Corning SM-332. Pump radiation from a continuous wave (CW) ytterbium-doped (Yb) fiber laser emitted at 1.125 μm was coupled through a 1.125/2.1 μm wavelength division multiplexer (WDM) to the ring cavity. Single stage isolator defined the direction of traveling wave. A commercial optical polarizer (OP) and a couple of polarization controllers (PC) were positioned between the active and SM fiber to form the NPE based fast light amplitude modulator. A polymer film with dispersed SWCNTs was used as a slow saturable absorber to ensure a self-starting mode-locking. A single-mode 50/50 fiber coupler was used as an output of the laser. So we have obtained an all-fiber holmium-doped laser hybrid mode-locked by co-action of NPE and SWCNT saturable absorber. Then an ultrashort pulse radiation from the MO was amplified by holmium-doped fiber amplifier. To suppress oscillation in the amplifier, an isolator was placed before a 1.125/2.1 μm WDM that combined the Yb-doped fiber pump laser emitted at 1125 nm and the pulses radiation being amplified.

The stable mode-locked operation was obtained at a central wavelength of 2076 nm with the 3 nm full width at half-maximum (FWHM) of the spectrum. The pulse repetition rate of 10.36 MHz corresponded to the cavity length of the MO being about 20 m. Figure 1(a) shows the pulse interferometric autocorrelation trace measured at a small gain of the MO radiation. The width of such pulses is 1.8 ps. Figure 1(b) shows the output spectra depending on the gain value. The presence of solitonic resonance sidebands is confirmed the negative cavity dispersion. It can be seen that spectra are deformed with the gain increasing. Such spectra dynamics can be related with the co-action of negative group velocities dispersion and nonlinear effects. The maximum obtained output power after amplification was about 244 mW that corresponded to gain about 25 dB with an input signal of 808 μW.

We have investigated the time and spectral dynamics of 2–μm ultrashort pulses changes during the amplification by holmium-doped fiber amplifier. The maximum obtained output power after amplification was about 244 mW that corresponded to gain value about 25 dB.

The research was carried out in the frames of the program of fundamental research of the Russian Academy of Sciences Presidium № I.7 "Actual problems of photonics, sensing of inhomogeneous media and materials".

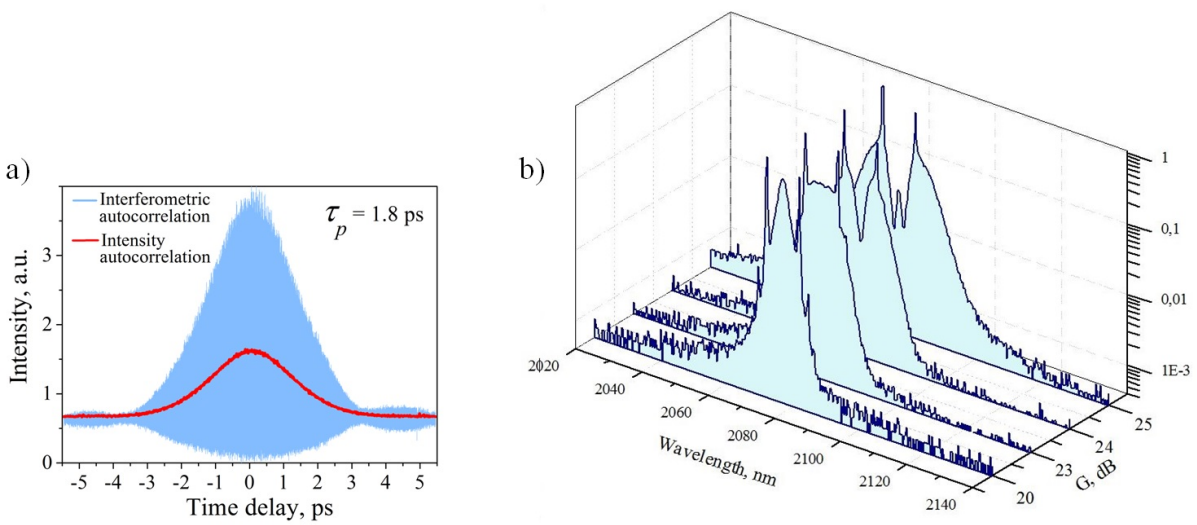

Figure 1: (a) - pulse interferometric autocorrelation trace, (b) - output spectra dynamics depending on the gain value.

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Towards chaotic generators of ultrashort light pulses

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Generators of chaotic sequences of pulses in the GHz range are in demand for cryptography and wideband communication [1]. We consider the prospects for pushing the regular and chaotic nonlinear laser dynamics into the time scale below a cavity round-trip time T_r, based on the idea of combining harmonic mode locking [2,3] with dual optoelectronic feedback [4], see Figure 1. Harmonic self-mode-locking, i.e. the generation of n>1 short equidistant pulses on a cavity round-trip time, is a self-starting process sustained by passive optoelectronic control in the absence of harmonics generator. The behavior of a solid-state laser in such regime was studied numerically in a wide gain range, including regular and chaotic nonlinear dynamics regions. Our main instrument is a detailed numerical simulation taking into account laser radiation fine time structure. We have shown that optimally chosen negative feedback delay $(1 - 1/n)T_\mathrm{r}$ leads to self-started mode-locking with generation of desired number of pulses n in the laser cavity. The second necessary condition for a self-mode-locking is the feedback inertiality which implies that the optimal value ($\sim T_\mathrm{r}/n$) of signal decay coefficient γ in a feedback control loop is large. This is a major obstacle in a way to independent chaotic generators in a laser cavity. To overcome it we introduced a second positive (T_r)-delayed feedback with the same value of γ. Such dual feedback control allows to switch off the interpulse interaction and to organize dynamics similar to n independent chaotic logistic maps coexisting in the laser cavity.

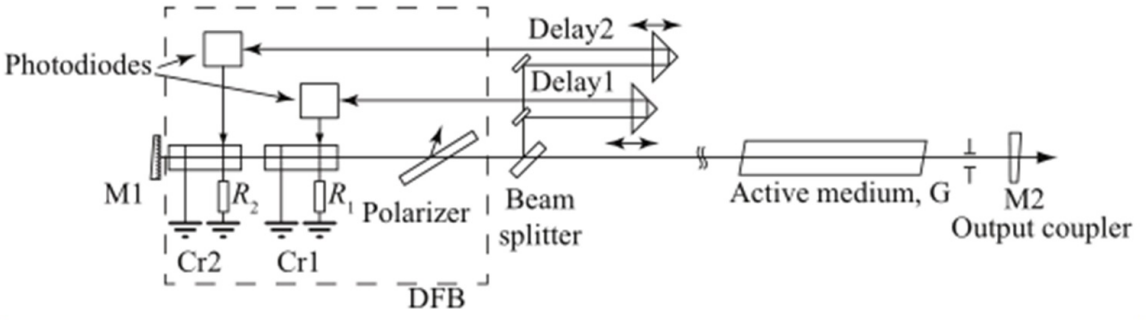

Figure 1: A simplified scheme of the laser controlled by two feedback loops based on two-sectional electro-optical modulator (crystals Cr1 and Cr2). M1, M2 – cavity mirrors, DFB —- dual–feedback optoelectronic control system.

To complement the detailed numerical simulation, we introduced a simpler approach based on point maps which allows one to reveal fundamental features of laser dynamics in the most illustrative way. The map for the case of combination of negative and delayed positive inertial feedback loops is

$$x_{n+1} = r x_{n-(k-1)}(1 - \sum_{i=0} x_{n-(k-1)-k\cdot M-i} \cdot \gamma^i + \alpha \sum_{i=p} x_{n-(k-1)-k\cdot M-i} \cdot \gamma^{i-p}), n = 0, 1, 2...,$$

where p is the delay in interpulse time intervals, α is the relative feedback sensitivity of positive feedback loop, x_n is the normalized energy of n^{th} ultrashort pulse in the sequence.

By choosing the value of α it is possible to cancel the members of the first sum and thus to switch off the feedback action. The proposed approach to laser control gives new possibility for ordering of output time structure and may be useful for a wide community of scientists and engineers that work in the areas of nonlinear dynamics, solid state lasers and optoelectronics.

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Nonlinear thermodynamics perspective of mode-locking

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Mode-locked lasers draw intense interest not only because they enable ultrafast optics, but also due to the rich physics of their dynamics. However, despite decades of efforts, there is no general theory of mode-locking. Here, we will describe a renewed perspective of mode-locking, particularly of fiber lasers, that has guided us (i) to formulate an algorithmic approach to designing specific mode-locked laser cavities that can produce desired pulse evolution characteristics and (ii) to predict how robust a mode-locked laser is against environmental perturbations. This is mostly a renewed, rather than new perspective, because the laser was among the first model systems of self-organization analyzed by H. Haken and others in the 60s and 70s, but this perspective appears to have been largely forgotten in recent decades.

Subpicosecond X-ray laser at λ=41.8 nm in a plasma formed by the interaction of a femtosecond pump laser with a xenon cluster jet

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Currently, free-electron X-ray lasers (XRL) with λ=1-10 nm and duration $t_{XRL} = 15$ fs allow holographic imaging of intermediate states of nano-objects for time comparable to atomic motion [1]. For many applications, compact XRL with short enough wavelengths and pulses shorter than 1 ps can be used. A principal requirement for XRL is the possibility of a one-shot pulse imaging, which implies emitted photon beam $\geq 10^{12}$ photon/pulse. XRL with the shortest amplification duration achieved to that time was demonstrated in [2-4].

Recently, a new class of XRL, based on optical field ionization of the krypton (Kr^{8+}, λ=32.8 nm [5-6]) and xenon (Xe^{8+}, λ=41.8 nm [7]) cluster jets is demonstrated. The approaches of these experiments can be used to develop ultra short XRL on the transition $4d_{5/2}5d_{5/2}$ [J=0] — $4d_{5/2}5p_{3/2}$ [J=1]. Prospects for developing the subpicosecond XRL is due to the anomalously high excitation cross-section of the upper active level $4d_{5/2}5d_{5/2}$ [J=0] by electron impact in Pd- like xenon (Xe^{8+}). The experiments [5-7] have shown the fundamental advantages of using cluster targets for creating highly efficient XRLs. These principal prerequisites were discussed in [8]: (i) Absorption of more than 90% of the pump energy is possible. (ii) Weak reflection of the pump beam from a cluster target and the absence of debris from plasma. (iii) Attainment of electron energies of several keV; energy deposition into cluster plasma, as well as ion and electron temperatures is controlled by the cluster size and the pump pulse contrast. (iv) Ionization balance is controlled by the pump laser intensity I_{pump}. (v) High purity and the absence of oxides.

The XRL calculation model was presented in our previous studies containing references to the detailed descriptions of the theoretical approach. Fig.1 demonstrates the comparison of the time dependences of the gains on the transition with λ=41.8 nm in Xe^{8+} in plasma at T_e=300 eV with experimental measurements [9]. (See Fig. 4 in [9]). In [9] plasma was produced by OFI of gaseous xenon. Gas pressures are 5, 10, 15, 20, 25 Torr (ion densities n_i= (1.75, 3.5, 5.25, 7.0, 8.25)$\times 10^{18}$ cm^{-3}). Some of the model calculations were performed in our work [10] before publication of the experimental data [9]. Thus, the reliability of our calculations is proved. Fig. 2 demonstrates the time evolution of gains that can be achieved by the interaction of the intense pump laser with the xenon cluster jet. We discuss the principles of achieving a quantum yield $>10^{12}$/pulse in a time $<$1 ps and experimental approach.

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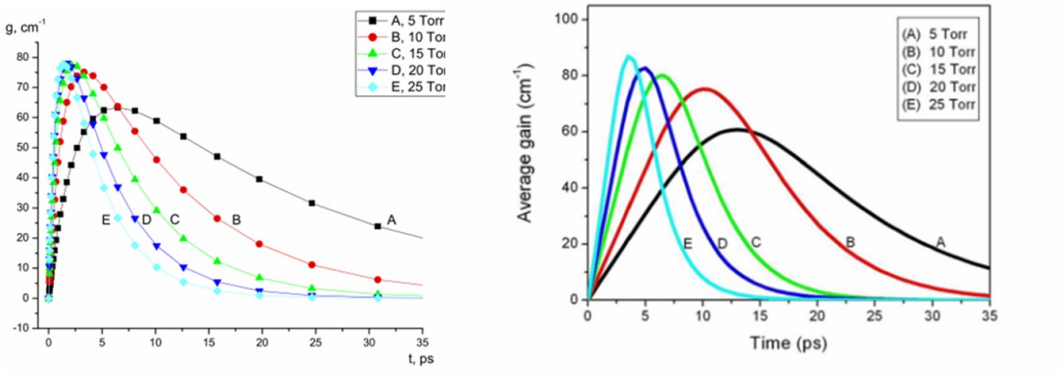

FIG. 4 (color online). Temporal profiles of the average gain coefficient extracted from the experimental data in Fig. 2 using the inversion procedure.

a) **b)**

Figure 1: Comparison of gain evolution on the transition $4d_{5/2}5d_{5/2}$ [J=0] — $4d_{5/2}5p_{3/2}$ [J=1] in Xe^{8+} between present calculation (a) gain values from Fig.4 of the experiment [9] (b).

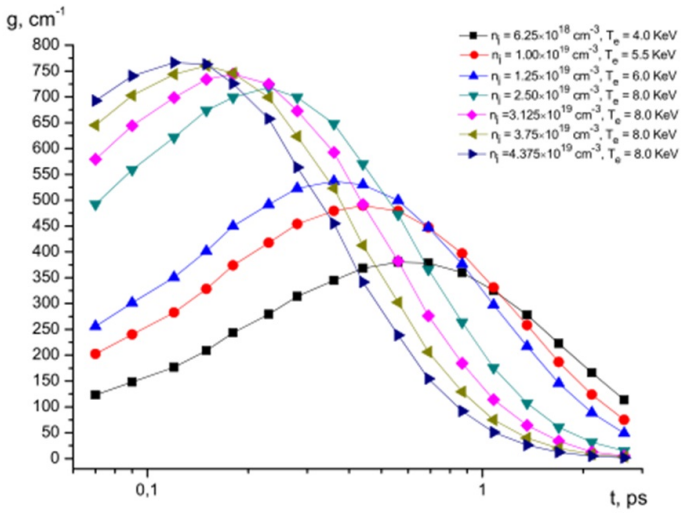

Figure 2: Time evolution of gains calculated at different ion densities and electron temperatures.

Compression after Compressor Approach (CafCA)

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We discuss different ways to use nonlinear optics in order to compress powerful laser pulse after a grating compressor. The basic idea is to use self-phase modulation and chirped mirror. The following aspects will be studied in detail: influence of residual spectral phase on self-phase modulation, two-stage CafCA, small-scale self-focusing suppression, second harmonic generation followed by CafCA, self-compression after compressor.).

Dielectric diffraction gratings for laser pulse compression

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The paper deals with the questions of development of holographic gratings with high radiation resistance for the laser pulse compression; as well it presents the attainments of JSC "NPO GIPO" for solution of this problem. Influence of technology parameters of dielectric diffraction gratings on spectral distribution of diffraction efficiency (DE) and damage threshold have been studied. Optimal technology parameters such as grating groove height, thickness of high- and low-refraction index interference layers and groove fill factor have been defined for various groove frequencies and incident angles (geometry parameters).

High-resolution acousto-optic ultrafast pulse shaping

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Acoustic-optic devices are widely applied in ultrafast optics as dispersive pulse shapers, frequency shifters, and pulse pickers. We report on state-of-art and latest advances in design and applications of quasicollinear acousto-optic dispersive delay lines in femtosecond laser systems. The results include but not limited to high-resolution time-domain pulse shaping, adaptive broadband dispersion control, and THz generation via optical rectification. Promising acousto-optical materials are observed as well.

High-peak-power diode-pumped picoseconds lasers

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Picosecond lasers with single pulse energy of mJ and multi-mJ level operating at moderate repetition rates up to $\sim$ kHz are required in a number of scientific and technological applications, such as time-resolved laser spectroscopy, ablation study, precise remote laser ranging, driving photoinjectors etc. Despite the peak power of the order of GW fractions, average power amounts only few watts and then the schemes based on diode pumped bulk crystals are most appropriate.

In this paper, we consider schematic solutions maintaining development of such kind of energy-effective, compact, robust and easy integrable picoseconds lasers based on Nd-doped crystals. We develop approach based on the dynamical operation control scheme utilizing pulsed repetitive operation regime, active and passive mode-locking, negative feedback, adjustable loss level in the oscillator cavity and switching regenerative amplification regime. This provides laser pulse formation in each laser shot and actually allows shortest way for obtaining just at the laser output near transform limited picosecond pulses of several millijoules energy with excellent pulse-to-pulse stability and low optical jitter value. Both oscillator and regenerative amplifier can be based on the same single active crystal. Using Fabry-Perot etalons inside oscillator cavity allows significant varying output pulse width which can take values from 15 (Nd:YLF) or 25 ps (Nd:YAG) and up to 300 ps.

Evolution of pulse energy, spectrum and time profile during a single generation cycle can be well illustrated using universal numerical calculation model which describes pulse formation governed by the operation control and also taking into account the pulse profile modifying due to amplification.

Utilizing diode end-pump geometry allows maximal overlapping of resonator mode and pumped volume, whereby providing optimal pump energy conversion efficiency into the output radiation. As a result, using the picosecond Nd:YAG laser based on the described scheme with one end-amplifier stage provides 25 ps pulses more than 5 mJ at fundamental wavelength with repetition rates within $\sim$400Hz. Conversion efficiency in the second harmonic with LBO nonlinear crystals is above 60%. Owing to pulse-repetitive regime and end-pump geometry, thermal loading is not high and the system does not require liquid cooling and can be easily power scalable by means of an additional amplification stage operating near the saturation regime.

At high repetition rates operation conditions strongly depend on thermal lens induced in the laser crystal. Increase of average end-pump power principally results in aberrational lens formation which may influence on mode locking and pulse amplification regime. Detailed experimental and modeling results are presented.

Generation of high intensity few-cycle femtosecond pulses from high power picosecond laser

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The increasing of repetition rate, optical efficiency and stability of high intensity lasers opens up for new opportunities of such systems in medicine, security and another applications. Combination of broadband OPCPA amplification, modern high average power pump sources (including high power ps lasers), the using of chirped mirrors and new types of nonlinear crystals greatly improves output parameters of modern high intensity lasers [1]. The developing of a high power picosecond laser system based on Yb doped thin rod and thin disk amplifiers and further OPCPA amplification by using this laser as a seed (via a white light generation) and pump sources are presented in this work.

The femtosecond fiber laser with 1 μJ of pulse energy at 3 MHz repetition rate is used as a seed source of laser system. The signal is stretched in chirped volume bragg grating (CVBG) from 300 fs to 0.5 ns pulse duration and amplified in two-stage Yb:YAG thin rod amplifier [2] up to 2 mJ at 11 kHz repetition rate and compressed to 1.1 ps in the same CVBG. The next amplification stage is a high power disk amplifier with up to 32 passes through the active element [3]. The output energy 11 mJ has been achieved with a near diffraction limited beam quality (M2 = 1.3) and the main limitation of output energy is residual phase distortions in disk active element. New disk active elements are already tested and its employment will allow increasing the output energy to $\sim$30 mJ at 11 kHz repetition rate. This hybrid laser system (with fiber, thin rod and thin disk active elements) provides a very efficient way to achieve both high energy and high average laser power.

The signal after thin rod amplification stages is used for generation of few-cycle femtosecond pulses. The compressed signal with 0.2 mJ of energy is converted to 515 nm and divided on 3 parts. The first part is focused to YAG crystal for white light generation. The red wing of this broadband radiation is parametrically amplified in BBO crystal by using another part of the signal as the pump. CEP stable idler in 1.5-2.5 μm spectral range with μJ level of pulse energy generates during this process. Additionally, the second harmonic of this IR signal is parametrically amplified to 2 μJ by using a third part of the signal as the pump source. The possibility of compressing of achieved radiation is investigated and less than 30 fs pulse duration (limited by measuring equipment) has been demostrated in both spectral ranges (1.5-2.5 μm and 0.7-1.0 μm). As the next parametrical stages, these two CEP-stabilized signals will be amplified to sub-TW intensity by using a 30 mJ picosecond laser at 11 kHz repetition rate as a pump source. Such tasks as influence of chirped parametrical pumping on conversion efficiency, non collinear parametrical amplification, ps to fs pulse conversion efficiency, CEP stability and others will be investigated under conditions of high average power and strong thermal effects.

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The deformation wave as a factor that controls direct laser writing

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We found a new regime of efficient laser modification of transparent materials based on the simultaneous actions of rarefaction and electronic excitation. It can be activated with a sub-nanosecond and nanosecond burst of femtosecond pulses. The scenario of the regime is following: a deformation wave is developed on a sub-nanosecond scale as a result of production of local excessive pressure by a tightly focused bullet of femtosecond laser pulse. Second and following pulses of the burst interact with the stretched material, and this condition translates to inscription of increased refractive index change in silica glass and sapphire.

Recent progress in mid-IR (4-5 μm) solid-state femtosecond amplifiers based on Fe^{2+}:ZnSe optically pumped by 3-μm laser

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Recent advance in development of powerful mid-IR lasers in 3-10 μm spectral range with ultrashort pulse duration opens up new horizons for investigating extreme laser matter interaction. Middle infrared femtosecond lasers offer a new paradigm for intense laser matter interaction mainly because ponderomotive electron energy scales as λ^2 [1]. A mastered way to create femtosecond mid-IR laser pulses with high peak power is to utilize optical parametric chirped pulse amplification scheme (OPCPA) [2]. Presently powerful OPCPA based GW systems with only 3.9 μm [2], 5 μm [3] and 7 μm [4] central wavelengths are available. However, filling the gap in between of 3.9 and 5 μm wavelengths seems to be very challenging due to the lack of suitable nonlinear crystals for parametric wavelength conversion. Indeed, oxide nonlinear crystals show transparency cutoff in this region, whereas non-oxide crystals could not be efficiently phase-matched. Iron-doped crystals offers a unique possibility to generate femtosecond pulses in the 3.5–6.1 μm spectral range [5,6].

The mid-IR seed pulses in our hybrid laser system originate from OPA consisting of three amplification stages. We have developed a robust optical parametric amplifier (OPA) based on three-$AgGaS_2$-crystal pumping by a Cr:Forsterite GW femtosecond laser system, generating 150 fs pulses in dual bands of 1.6—2.0 μm (signal wave) and 3.5-–5.5 μm (idler wave). By introducing a negative prechirp to the pump, a combined efficiency in two waves of greater than 10% was achieved, with signal energy up to 110 μJ and idler energy up to 43 μJ [7]. Further amplification stage in multi-pass Fe^{2+}:ZnSe amplifier optically pumped by a solid-state Q-switched Cr:Yb:Ho:YSGG laser (30 mJ, 2.85 mm, 5 Hz) with output energy up to 2.1 mJ. Nonlinear compression in bulk CaF_2 [8] due to the SPM and anomalous GVD provides the enhancement of peak power up to 20 GW. In order to reach sub-TW or even TW power level the gain medium pumping area should be 2-3 cm^2 which requires high energy pump to be used. In order to maintain the optimal pump energy density of about 0.5–1 J/cm^2 2–3 J of pump energy is needed.

By utilizing high energy pump technology the Fe^{2+}:ZnSe laser systems have the capability for scaling to higher energies (up to 200 mJ) and peak powers even up to TW through the incorporation of additional amplification stages using total pump energy of 2–3 J and nonlinear dielectric compressor installed at the front end of the mid IR femtosecond laser system. Such an energy of pump is achievable for non-chain electro-dischargeable HF laser operating in pulse-periodic regime.

We believe, that principles which form the basis to create gigawatt power level femtosecond laser system based on Fe^{2+}:ZnSe optically pumped by solid-state Q-switched 3 μm, can be applied to the development of femtosecond powerful lasers operating over the mid-IR spectral range from 3.5 μm (Fe^{2+}:ZnS) up to 6.1 μm (Fe^{2+}:CdSe, Fe^{2+}:CdTe) [9].

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kW-class sub-1-picosecond lasers followed by efficient UV and mid-IR frequency conversion for laser-matter interaction research

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Thin disk lasers operating in near infrared spectral range are powerful tools for generation of kW beams in temporal range from CW to ultrashort pulses. Especially picosecond disk lasers have become indispensable instruments in many industrial processes like accurate drilling of tiny holes, multi-beam large area surface micro- and nano-patterning, or plasma creation for incoherent extreme UV and soft x-rays generation. However, state-of-the-art industrial applications are pushing laser development to generation of shorter pulses, higher average power, and broadening number of available wavelengths ranging from UV to mid-infrared. As a consequence, high power pulsed laser systems based on Yb:YAG have got booster amplifiers, new active laser materials for mid-infrared light generation have been adopted, or thin disk laser pumped frequency conversion stages have been constructed.

Field of interest of the Hilase centre in Dolni Brezany (Czechia) are picosecond and sub-picosecond lasers in near- and mid-infrared followed by frequency conversion to UV and visible spectral regions. We routinely use a 500-W laser called Perla C which delivers up to 5 mJ (100 kHz) or 10 mJ pulses (50 kHz) in a nearly diffraction limited beam ($M^2 < 1.8$) and sub-2-ps long pulses [1]. The system is based on an Yb:YAG (1030 nm) ring cavity regenerative amplifier and uses a unique, compact pulse compressor based on a large chirped volume Bragg grating. Besides, the system integrates a unique in-house developed Pockel's cell based on large aperture BBO crystal, and the active medium is efficiently pumped at 969 nm directly to upper laser level (so called zero phonon line) instead of widely used wavelength of 940nm. This brings lower thermal load and higher achievable output power. Upgrade of the system up to 1 kW is under design now. We target on boosting the amplifier by adding one more gain medium to its cavity.

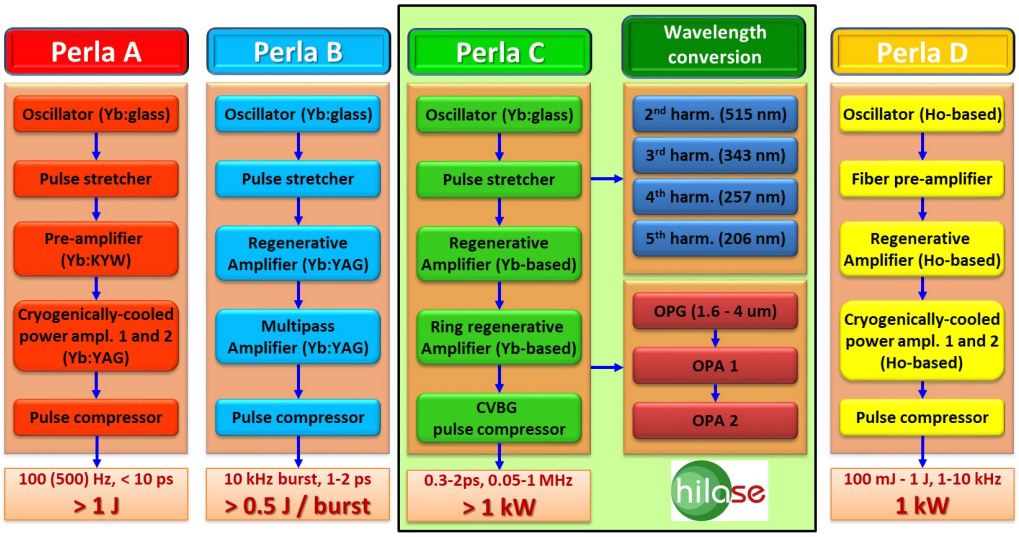
Frequency conversion to 2^{nd}, 4^{th}, and 5^{th} harmonic frequency from the Perla C was realized [1,2]. Up to now, we pumped the conversion system by less than 70 W. Conversion efficiency from fundamental to 2^{nd} harmonic approaching 60%, 18% to 4^{th} harmonic, and <2% to 5^{th} harmonic in sub-1-ps pulses was realized. At 257.5 nm we reached one of the best results published in picosecond regime by that time [2]. Conversion to longer wavelengths is designed like a PPLN-based OPG followed by parametric amplification in a KTA or a KTP crystals. We repeatedly reached tunability from 1.46 to 1.95 um (signal beam) and 2.18 – 3.5 μm (idler beam) with total average power of 14 W (42 W pump) and < 2 ps pulse length [1]. We aim to reaching average power of 100 W in this spectral region. For OPA at longer wavelengths we plan to use a new femtosecond Ho:YAG based thin disk beamline targeting to 100 W, or to apply difference frequency generation (DFG) in semiconductor materials after amplification of pulses generated close to 2 μm. The systems are under design.

Apart from the Perla C, high energy Yb:YAG laser systems Perla A and Perla B are under development. Perla B is designed like a CPA (chirped pulse amplification) system consisting of a 45 W thin disk regenerative amplifier followed by a multipass amplifier

and a grating compressor delivering sub-1-ps long pulses in a 10-kHz-burst with burst repetition rate of 100 Hz. First phase is designed for 500 mJ bursts, later multi-Joule-bursts will be realized by scaling of the thin disks and adding and more multipass amplifier. The Perla B front-end is also capable of continuous pulsing with repetition rate of 1 kHz up to 45 W of average output power [3]. Perla B conversion stages for harmonic frequency generation are under design.

High energy laser system Perla A is focused on generation of 1-J picosecond pulses. To store and extract efficiently high pulse energy from a disk-shaped gain medium, a concept of a cryogenically cooled Yb:YAG amplifier was chosen. Lately we demonstrated generation of 103 mJ picosecond pulses (bandwidth 0.4 nm) stretched to 0.5 ns at repetition rate of 100 Hz. After adding a liquid nitrogen-cooled power amplifier, generation of short pulses up to 1 J of pulse energy is expected. Lately we have started also development of sub-picosecond multi-100-W lasers as requested by application projects at the Hilase centre. We are directing this research in two directions – application of new broadband materials and spectral broadening of picosecond pulses in gas cells. In preliminary experiments we managed to directly generate 300 fs long pulses from Yb:YGAG ceramic material, which has similar thermomechanical properties to Yb:YAG but broader bandwidth [4]. This year a second phase, demonstration of a femtosecond high power disk laser is expected.

The latest results of the group of picosecond disk lasers will be presented at the conference.



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Stretchers and compressors of chirped pulses — key elements of ultra-high-power laser complexes

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Rapid progress is currently observed in the creation of petawatt and multipetawatt laser systems. These systems have unique properties: unprecedented values of intensities, unattainable on the Earth by other methods, comparable to light intensities in the interior of stars may be obtained by means of focused radiation. This makes such sources indispensable for fundamental research, laboratory astrophysics, power engineering, biology, medicine, and other branches of science and technology. Record values of peak power achieved at the present are about 5 PW.

Despite a large diversity of high-power laser complexes worldwide, all of them are based on CPA or OPCPA – chirped pulse amplification or optical parametric CPA. The basic idea underlying the CPA (OPCPA) technique is amplification of an ultrashort pulse pre-stretched in time by 3–4 orders of magnitude and its subsequent compression to the initial duration. Pulse stretching greatly decreases its peak power, thereby allowing the pulse to pass through the amplifying medium without unwanted self-focusing effects.

Thus, stretchers and compressors are undoubtedly the key elements of high-power laser CPA/OPCPA complexes. The stretchers are used for stretching ultrashort pulses in time, and the compressors for compressing the amplified chirped pulses. The spectral components of broadband radiation that has passed through the stretcher lag behind one another due to dispersion, as a result of which the pulse is phase modulated (chirped) and, at the same time, stretched in time. The sign of the group velocity dispersion in the compressor is opposite to that of the stretcher dispersion. Therefore, the corresponding spectral components in the compressor overtake each other and the pulse compresses to the duration close to the initial one.

For over than a 30-year period from the appearance of the first CPA system, quite a number of various stretches and compressors have been created. Basic schemes of these devices, as well as requirements to accuracy of their adjustment and matching of dispersion characteristics are considered. With the advance of the CPA/OPCPA systems, matching of higher dispersion orders is becoming an increasingly more important problem. It is especially significant for present-day high-power laser systems having pulse duration less than 30 fs.

Great attention is devoted to the background of the CPA and OPCPA systems, with particular emphasis on the history of the ideas of creating stretchers and state-of-the art compressors showing the elegance and beauty of these devices.

Parametric Amplification of Few-cycle Optical Pulses

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Section 7: Femtosecond radiation in spectroscopy and optical frequency metrology

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Scope
Absolute optical frequency measurements
Femtosecond frequency combs for direct spectroscopy of ions and atoms
UV and HUV frequency combs
Frequency combs in astrophysics
Optical frequency combs applications

Characterization of 1.14 μm clock laser by Ti:Sa femtosecond frequency comb

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Our group is working on thulium optical lattice clock based on the inner-shell transition at 1.14 μm. In previous work, we have shown that the clock transition have low sensitivity to blackbody radiation shift and the total relative frequency uncertainty below 10^{-17} could be reached [1].

By now we have performed a spectroscopy of the clock transition in the optical lattice at the magic wavelength and obtained a spectrum with 10 Hz width. As a clock laser we use semiconductor Thoptica DL-Pro laser which is stabilized to external high-finesse ULE cavity by Pound-Drever-Hall technique. This technique provides the relative frequency instability of smaller than 10^{-14} in $1-100$ s integration time [2]. The clock laser frequency is measured by femtosecond frequency comb. We use an optical frequency comb with a repetition rate of 985 MHz, which is locked to a passive H-maser continuously calibrated to GPS. Measurement of the clock laser frequency during a week have shown a linear drift of 29 mHz/s caused by ULE-glass aging. The linear drift is compensated by acousto-optical modulator. After compensation, the clock laser can be used as a stable frequency reference with residual variations on the level of 1 Hz at the timescale of 1000 s.

By measuring clock laser frequency during clock transition spectroscopy we determined the absolute frequency of clock transition in Tm $[\mathrm{Xe}]4f^{13}6s^2(J=7/2, F=4) \rightarrow [\mathrm{Xe}]4f^{13}6s^2(J=5/2, F=3)$ of $\nu = 262954938269213(30)$ Hz. The frequency uncertainty mainly comes from instability and calibration accuracy of a GPS-disciplined H-maser.

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Tailored nonlinear optical frequency conversion - toward high resolution spectroscopy in the vacuum ultraviolet wavelength region

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Nonlinear optical process is strongly dominated by phase relationships among the electromagnetic fields relevant to its optical process. If we can manipulate this phase relationships to arbitrary values at arbitrary interaction lengths, we can tailor the nonlinear optical phenomenon to a variety of ways.

As a typical example, we applied this concept to the Raman-resonant four-wave-mixing process in gaseous parahydrogen [1]. We showed near-unity quantum-efficiency frequency conversion to an aimed high-order Stokes or anti-Stokes component on the basis of detail numerical simulation. In this study, we have also shown that a single-frequency tunable laser which covers an entire wavelength region of 120 – 200 nm in the vacuum ultraviolet could be realized [1]. Here, to manipulate the phase relationships among many electromagnetic fields, we employed a method which precisely control thickness of a transparent dispersive plate [2,3,4]. We could also find that this technology could function very practically through this study.

Fig. 1 is a typical result demonstrated in reality which well reproduced the prediction of the numerical simulation [5,6]. Generation of 1st anti-Stokes was enhanced or suppressed depending on the phase relationships, that is, by controlling thicknesses of the inserted dispersive plates. White color in fig. 1b expresses that the anti-Stokes component was strongly generated.

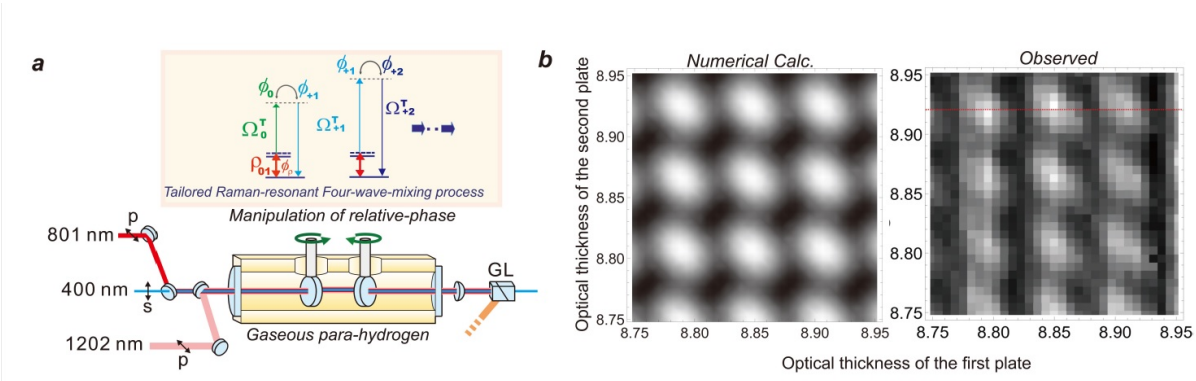

Figure 1: Amplitudes/phases manipulation and the waveform obtained.

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Broadband absorption and dispersion spectroscopy with optical frequency comb

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I will present recent developments of the comb-based cavity-enhanced broadband absorption and dispersion spectroscopy. The comb-line resolving approaches and Fourier transform spectroscopy with sub-nominal resolution overcome the frequency resolution limits of conventional techniques. By the measurements of the high-finesse cavity mode's shapes we test the limits of comb-based techniques and apply it for absorption and dispersion measurements based on cavity mode width and shifts, respectively. Description of the possible detection schemes based both on dispersive elements and Fourier Transform will be given and advantages for various applications will be discussed.

Direct frequency comb two-photon spectroscopy and the proton radius puzzle

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The optical frequency comb, highlighted by the 2005 Nobel Prize in physics, allows direct comparison of optical frequencies with each other and with radio frequency references. In combination with ultra-stable CW lasers this led to a rapid progress in the field of high-precision atomic spectroscopy. Direct frequency comb spectroscopy of atomic transitions is an interesting alternative for transitions in the UV region, as pulsed lasers are readily up-converted. Doppler-free spectroscopy techniques, like two-photon transitions, are required in those experiments to mitigate systematic shifts due to the motion of the atoms.

We report preliminary results of direct two-photon frequency comb spectroscopy of the 1S-3S transition in atomic hydrogen and deuterium. We discuss systematic shifts, specific for this method together with further prospects of this experimental technique.

Combining our results with the well known value of 1S-2S transition frequency, one can extract the proton charge radius and the Rydberg constant. This is an important contribution to the so-called proton radius puzzle, that summarized a large disagreement between values of the proton charge radius obtained from various spectroscopic and electron-proton scattering experiments.

Optical frequency comb applications for large missions

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Optical frequency comb has been originally invented for the precision frequency ruler at the optical region, and it has been applied for various kind of fields such as ultrafast spectroscopy, OCT, astronomy etc. We have applied an EOM-based optical frequency comb to the timing synchronization for the 1-km X-ray free electron laser. In the frequency domain, the EOM-based optical frequency comb is used as the synchronization of a series of rf accelerators. And, in the temporal domain, it also used as the timing synchronization of X-ray pulses. Combination with the phase stabilized fiber network, high precision timing system would be realized. We will also present the space borne stable microwave generator based on a robust mode-lock optical fiber comb iand an iodine-stabilized Yb-doped fiber laser.

State of the art precision metrology with ultra-low-noise optical frequency combs

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The ongoing improvements on optical frequency standards based on cold atoms trapped in a lattice and ultra-stable cavity-stabilized lasers require a continuous advancement for optical frequency combs that are used as clockwork.

Recently, a laser source with a breakthrough 10 mHz linewidth, representing a coherence time above 10 seconds, and with very low frequency drift has been demonstrated [1]. Cryogenically cooled optical frequency standards based on Strontium atoms trapped in lattice have been demonstrated at 10^{-18} level of accuracy and stability. Moreover, the first Fermi-degenerate three-dimensional optical lattice frequency standard has been reported [2], showing the possibility to overcome the tradeoff between low quantum projection noise limit and density-dependent frequency shifts. For technical reasons, the operating frequency of the novel low-noise reference lasers is centered at approximately 194 THz, while the clock transition of neutral Strontium atoms is located around 429 THz. The Menlo Systems figure 9® Frequency Comb [3] acts as a clockwork capable of transferring the spectral purity of a reference laser at 1542 nm to the interrogation wavelength of a Strontium clock, paving the way for a novel level of instability and inaccuracy at 10^{-19} in optical frequency measurements. Moreover, the principle of spectral purity transfer can also be used to generate low-noise microwave signals. These are required for many applications such as a fly-wheel for microwave atomic clocks, coherent radar systems or deep-space communication. Such low noise microwave signal can be generated using a photonic microwave synthesizer in which an optical frequency comb is used as an optical frequency divider to phase coherently divide down an optical reference to the microwave domain. With appropriate optical and photodetection techniques, unrivalled ultra-low noise microwave signals can be generated [4].

Overall, the Frequency Comb has proven to serve as a spectral purity divider from optical lattice clock frequencies (429 THz), over 4 decades, down to the microwave regime ($\sim$12 GHz).

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Femtosecond laser synthesizer for methane reference oscillator

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Compact and reliable microwave reference oscillators with (1-5)$\times 10^{-15}$ (for averaging times τ = 1–1000 s) instability and low phase noise are needed for variety of applications — frequency etalons based on atomic fountains, radio astronomy, radars, etc [1]. Common approach to their development is based on frequency stabilized lasers and further optical-to-microwave frequency division (OFD) [2]. To divide the frequency from the optical range into the radio band, a femtosecond system based on a fiber laser is usually used.

At present, we have created an optical frequency synthesizer for time-frequency metrology based on a femtosecond erbium fiber laser and a variety of stabilizing frequency parameters.

The synthesizer is based on all-PM fibers. The synthesizer oscillator has two feedback loops for locking to a reference oscillator: a fast loop - electro-optical phase modulator and a slow loop - piezo-ceramic actuator. To stabilize the offset frequency synthesizer can be equipped with an f-2f interferometer.

In addition, several offset free schemes were implemented with generation of a difference frequency at 1550 and 3390 nm. The schemes include an optical frequency comb based on a Er fiber femtosecond laser and the generation of a difference frequency in a nonlinear crystal PPLN where only one parameter - the repetition frequency of the comb pulses must be stabilized [3].

At present, according to the scheme of difference frequency generation, two reference oscillators based on a femtosecond fiber laser ($\lambda = 1.55$ μm) optical to microwave frequency divider and He-Ne / CH_4 optical frequency standard have been created. Preliminary measurements demonstrated a short-term instability of the output microwave signal at 1.5 GHz carrier frequency at a level of 1×10^{-14} ($\tau = 1$ s). The interrogative oscillator with such stability satisfies the requirements of application in atomic Cs and Rb fountains.

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Arbitrary optical waveform at 125THz repetition rate and its application to ultrafast phenomena

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Arbitrary generation of a continuous optical waveform at over 100THz repetition rate has been achieved. We can generate a series of phase-locked highly discrete harmonics by optical frequency division technique and control the spectral amplitudes and phases to get a target waveform in time domain. In this speak, we would like to discuss the arbitrary optical waveform generation and its potential in the ultrafast physics.

Arbitrary waveform generation system consists of three functional sections; firstly, optical frequency division section, secondly, amplitudes and phases manipulation section, and lastly, detection section. In the first section, five harmonics (1f-5f, f = 125 THz) are generated from two single frequency lasers (2f & 3f) and phase-locked using PPLNs. [1] CEP is also fixed. In the second section, amplitudes and phases of the five harmonics are manipulated by changing thicknesses of dispersive media on the optical path. [2] The five harmonics are not separated and stay coaxial throughout manipulations. This coaxial feature makes the system robust to external perturbations. In the last section, five amplitudes are measured, meanwhile phases are retrieved by detecting autocorrelation waveforms. From the information obtained, the target waveform can be constructed.

Fig. 1 shows a generated saw-tooth optical waveform at 125 THz repetition rate. The waveform is almost congruent with a Fourier-limited one. Phase stability has been confirmed around 1 hour by monitoring autocorrelation waveforms.

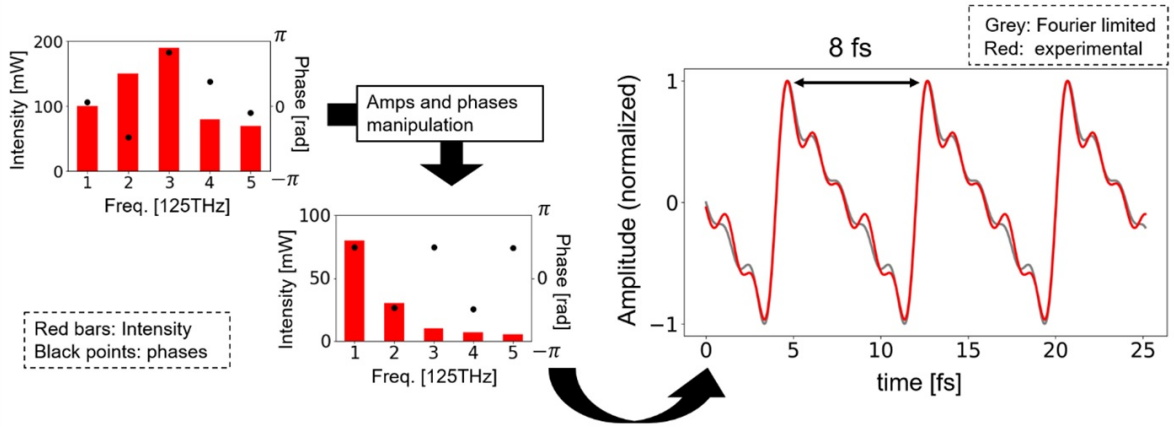

Figure 1: Amplitudes/phases manipulation and the waveform obtained.

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Generation of ultrashort pulses by arbitrary manipulating amplitudes and phases

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We report a new method of manipulating the amplitudes and phases of highly-discrete optical spectrum [1,2]. One of the applications of this technique, a generation of ultrashort pulses, has been demonstrated.

The initial coherent discrete spectrum with frequency spacing of 125 THz was produced through a stimulated vibrational Raman scattering in parahydrogen. First, in order to achieve nearly uniform amplitude distribution, we used a waveplate with varying thickness and a polarizer. Second, for the phase manipulation, we used two different dispersive plates (fused silica and calcium floride). Changing thickness of these two plates allowed us to arbitrarily change phase distribution between six components of the spectrum that we took into account. As a result, we were able to achieve ultrashort pulses with a duration of 1.4 fs and a repetition rate of 125 THz. SPIDER technique [3] was used to characterise the pulse.

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Kerr combs and solitons from regular diode lasers

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Over the past two decades optical Kerr frequency combs in high-Q optical microresonators [1] have found a wide variety of applications, such as coherent communications [2], dual-comb spectroscopy [3], astrophysical spectrometer calibration [4], LIDAR [5] and optical frequency synthesis [6]. The possibility of mode-locking via generation of dissipative Kerr solitons (DKS) has been demonstrated on a variety of platforms [7-10]. Especially DKS has provided broadband low-noise states with high repetition rates in the microwave to terahertz domain (10 GHz – 1 THz). Such combs may be packaged in a compact comb source with low power consumption. For obtaining DKS in microresonators a tunable ultra-narrow linewidth laser is traditionally used for pumping. However, the same type microresonators could be used for significant linewidth narrowing of laser diode exploiting resonant Rayleigh backscattering [11] for self-injection locking [12]. Previously for self-injection locking only single frequency prestabilized laser diodes were used with either Bragg-grating [12] or distributed feedback configuration [10] with narrow linewidth comparable to the resonance linewidth of the high-Q microresonator.

Here we demonstrate that initial stabilization is not required for soliton comb generation, so simple commercial broadband laser diodes may be used for comb generation in compact packaging device with low-power consumption. The different platforms, MgF_2 crystalline microdisk and Si_3N_4 integrated microring, are used for microresonator manufacturing. Self-injection locking to the microresonator with high Q-factor allows to significantly narrow the laser diode linewidth. The role of the microresonator is twofold: 1) it selects and narrows the linewidth of the laser diode via self-injection locking, and 2) Kerr comb is generated in the microresonator. We use the multi-frequency Fabry-Pèrot InP laser diodes (Seminex, central wavelength $\sim$ 1535, 1550 and 1650 nm; spectrum width $\sim$ 10 nm; output optical power $\sim$ 100 mW). The self-injection locking to the microresonators leads to the narrowing of the locked laser mode and the suppression of other longitudinal laser modes. Thus, the multi-frequency laser diode is converted into a single frequency laser source. The engineered group velocity dispersion (GVD) and the spontaneous four-wave mixing lead to the Kerr-frequency comb observation. Generation of soliton Kerr combs (beat note linewidth <1 kHz and spanning >25 nm) for the MgF_2 microresonator and wide Kerr comb (spanning >300 nm) for the Si_3N_4 microresonator is observed when the laser current was adjusted. This novel method allows to generate stable comb states in different platforms of microresonators by a commercial broadband multi-frequency laser diode without any stabilization (comb stability $\sim$ several hours). This result leads to a development of compact, low-noise soliton Kerr comb sources that are significantly important for wide range of applications.

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Yb:KYW frequency comb for precision spectroscopy of 1s-2s transition in He^+

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Precision spectroscopy of simple atomic systems like a hydrogen atom or a singly ionized helium are of a great importance for modern physics. As transition frequencies in these systems can be calculated ab initio and can be expressed through fundamental constants such as fine structure constant, Rydberg constant and proton charge radius, these measurements allow precise evaluation of these values for metrological purposes. On the other hand these experiments can be utilized as extremely sensitive tests of fundamental theories such as QED. At the moment an experiment aimed for a precision spectroscopy of He^+ is being in progress at Max Planck Institute for Quantum Optics in Germany. Transition frequencies in He^+ are more sensitive to high-order QED corrections than transitions in hydrogen atom which potentially allows more accurate verification of the theory. Spectroscopy will be performed with a frequency comb centered at 60 nm which will be obtained from a near-infrared comb via high-harmonic generation, which sets stringent requirements on a seed frequency comb in terms of spectral noise due to carrier collapse effect. Here a current progress on an Yb:KYW solid-state frequency comb designed for this experiment will be presented as well as a status of the whole experiment.

Author Index

Acoustic diagnostics of multiple and superfilamentation under different crossing angles between the separate filaments

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In this paper we studied the superfilamentation of high-power femtosecond laser radiation using lenses with different focal lengths. Radiation of several filaments merge near the lens focus and a superfilament is formed – a filament, that contains much more intensity and electron density than a common one [1].

In our experiments we used laser system, based on Ti:Sa crystal (805 nm, 1-10 Hz, beam diameter (FWHM) 7 mm, energy up to 20 mJ, pulse duration 55 fs) to form filaments. The filament was created in air by different lenses (focal lengths from 20 cm to 300 cm). Regular structure of the filament was set by an amplitude mask – opaque plate with four holes located at the vertices of a square.

For the filament diagnostics we used two independent methods simultaneously. Firstly, it was acoustic method. A broadband piezoelectric transducer contains a 110 μm thick polyvinylidene fluoride (PVDF) film with 6 mm working area diameter. The signal from the piezoelectric transducer was amplified by a broadband amplifier with a gain of 12 and recorded on a computer. Working bandwidth of the transducer was up to 6 MHz. Secondly, the mode of radiation in the filament was registered. A silica wedge was placed in 10 cm away from the piezoelectric transducer reflecting $\sim$ 4% of radiation from the filamentation zone. An image from the wedge surface transferred to the CCD camera. The ImagingSource dmk 23fv024 camera with 752x480 matrix and 6 μm pixel size was used. The wedge was shifted after each shot so that the reflection occurred from a clean surface.

The acoustic method was used for filament parameters estimation. Measuring the amplitude and width of an acoustic signal it is possible to estimate the size of the heat source and the volume density of the absorbed energy. Linear energy density was calculated using a formula form two previous parameters.

In the case of filament formed by a 3 m lens, a nonlinear increase in the linear energy density with an increase in the number of merging filaments was observed – a superfilament formation. The fusion occurred less efficiently with a decrease in the focal length of the lens. It is explained by the large convergence angles between individual filaments. A central filament formation in the case of large convergence angles is explained by the constructive interference of regularized filaments.

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Giant optical fields in dielectric resonance metasurfaces and SERS effect

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Anomalous optical response of metasurfaces, composed of silicon regular resonators is investigated. The resonators have form of two-dimensional bars or regular cones and covered by semicontinuous gold or silver films. Experiments and computer simulations demonstrate excitation of multiple metal-dielectric resonances. For example, the silicon cone resonator exhibits various em modes including dipolar modes and hybrid whispering gallery modes. The axial symmetry of the cone results in the angular momentum quantization and excitation of the modes with polar quantum number $l \gg 1$, azimuthal quantum number m, and radial quantum number q. The highest quality factor corresponds to the modes with minimal l and q but large m. The local electromagnetic field is greatly enhanced in-between metal particles placed on the surface of the dielectric resonators.

The electric field enhancement and surface enhanced Raman scattering (SERS) in periodic dielectric metafilms was obtained in [1-3] for microwave and optical spectral ranges. The optical metafilms with top metallization comprising of 20 nm thick silver layer were also considered. The anomalous optical response from a metasurface is investigated by measuring the reflection for various angles of the light incidence. In [4] we investigated the metasurface, composed of silicon nanocones that reveals extraordinary optical diffraction and SERS effect. We also suggest concentrating the optical field at the vertex of a dielectric beak, which is attached to the dielectric resonator [5]. The spatial scale of the hot spot, where the field concentrates, is determined by the curvature of the vertex and can be less than a nanometer. The obtained results open up new possibilities for R&D the SERS substrates including sensors for detecting specific substances.

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Femtosecond annular light bullets in a medium with anomalous group velocity dispersion

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Conception of light bullets (LB) considers a consistent compression of the laser pulse in space and time with high localization of the light field. Light bullet is a robust, but short-lived formation with peak intensity exceeding 10^{13} W/cm^2. [1]. In bulk transparent dielectric, the formation of bullets during the filamentation of a femtosecond laser pulse was numerically studied in [2, 3]. The formation of the LB sequence predicted in [3], was confirmed experimentally in [4, 5] where the dynamics of LB along the filament in fused silica was investigated.

Annular beams with a spiral phase dislocation on the optical axis (optical vortices) preserve the circular structure with on axis zero field in nonlinear medium. The phase dislocation influence on a spatiotemporal dynamics of the powerful femtosecond annular pulsed beam at the wavelength 800 nm in fused silica was numerically studied in [6]. It is shown that optical vortex in a medium with normal group velocity dispersion forms a filament of tubular shape with a radius of several microns, whereas in an annular beam without phase dislocation, the filament is formed on the axis.

This work is devoted to studying the spatiotemporal compression of a femtosecond annular beam with phase dislocation in presence of anomalous group velocity dispersion at the wavelength of 1800 nm in fused silica.

Numerical simulation of the femtosecond pulse propagation is based on the system of equations for slowly varying complex amplitude of laser field $A(r,t,z)exp(im\phi)$ and free electron concentration $N_e(r,t)$:

$$\begin{cases} 2ik_0\dfrac{\partial A}{\partial z} = \hat{T}^{-1}\left[\left(\dfrac{1}{r}\dfrac{\partial}{\partial r}\left(r\dfrac{\partial}{\partial r}\right) - \dfrac{m^2}{r^2}\right)A\right] + \hat{T}^{-1}\hat{D}[A] + \dfrac{2k_0^2}{n_0}\hat{T}\Delta n_k A - \dfrac{2k_0^2}{n_0}\hat{T}^{-1}\Delta n_{pl}A + \\ \qquad +i\hat{T}^{-2}\sigma A - ik_0(\alpha+\delta)A, \\ \dfrac{\partial N_e}{\partial t} = R_E(N_0 - N_e) + N_e(\nu_i - \beta). \end{cases}$$

The propagation equation includes operator of wave non-stationarity $\hat{T}$.

The complex envelope amplitude of the electric field in optical vortex at the beginning of the propagation distance (at $z = 0$) is given by:

$$A^{(m)}(r,\phi,t,z) = A_0\left(\frac{r}{r_0}\right)^m \exp\left(-\frac{r^2}{2r_0^2}\right) \exp\left(-\frac{t^2}{2t_0^2}\right)\exp\left(im\phi\right).$$

Pulsed beam parameters were chosen as $m = 1$, $r_0 = 100$ μm, $t_0 = 36$ fs, $\lambda_0 = 1800$ nm, $P = 5P_{cr}^{(1)}$, where $P_{cr}^{(1)}$ is a critical power for optical vortex with topological charge $m = 1$ [6], initial peak intensity $I_{max0} = 2.3 \times 10^{11}$ W/cm^2 and energy $E_0 = 12.7$ μJ. Parameter of anomalous group velocity dispersion $k_2 = \partial^2 k/\partial\omega^2|_{\omega=\omega_0} = -62.6$ fs^2/mm.

The Kerr self-focusing of femtosecond optical vortex together with pulse compression leads to the formation of a sequence of several annular light bullets with peak intensity up to 5×10^{13} W/cm^2. The first one is formed at a distance of $z = 0.9$ cm. (Fig.1). This and subsequent light bullets are shifted in time towards the tail of the pulse due to the

delayed Kerr nonlinearity, as well as the nonlinear increase in the refractive index and, consequently, the decrease in the group velocity.

The radius of the consecutive annular structures decreases from bullet to bullet up to $5\lambda_0$. The duration of wave packets in each bullet remains approximately the same and is equal to 10 fs, which is 1-2 optical field oscillations.

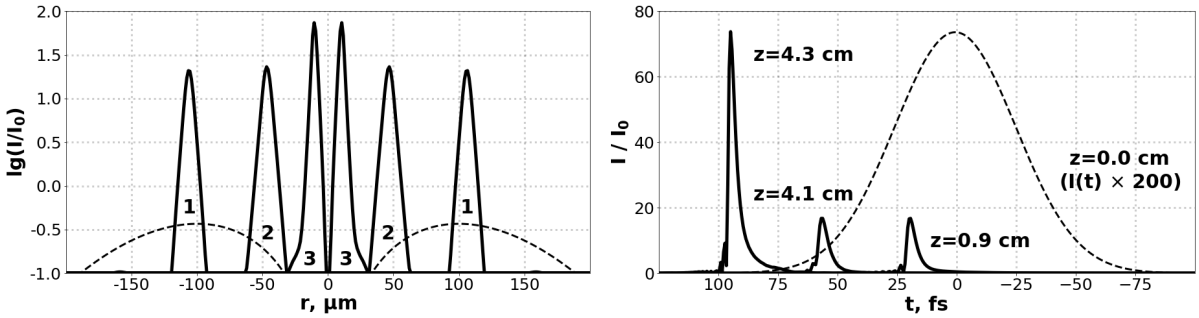

Figure 1: Spatial (left) and time (right) profiles of light bullets at distances: $z = 0.9$ cm (1), 4.1 cm (2) and 4.3 cm (3). Initial profiles are denoted by dashed lines (intensity in time profile is multiplied by factor 200). Spatial profiles are presented in logarithmic scale.

The energy of light bullets is 5-25% of the energy of the initial pulse. The decrease of energy in the sequence of bullets is associated with its scattering to the background, as well as with a complex spatiotemporal transformation of the pulse, at which a certain amount of energy flows to the periphery of the beam.

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