Book of Abstracts

V International Conference
on Ultrafast Optical Science

UltrafastLight-2021

October 4 – 8, 2021
Lebedev Physical Institute, Moscow

V International Conference on Ultrafast Optical Science “UltrafastLight-2021”, is the broad-scope, annual international symposium dedicated to the most important aspects of ultrafast phenomena in different fields of natural sciences and engineering.

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1. Extreme light
2. Ultrafast phenomena in ionized gases
3. Ultrafast phenomena in condensed matter
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5. Frequency combs in spectroscopy and optical clocks
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7. Ultrafast laser technologies in microoptics, nanophotonics and structured light
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Scope

Laser particle acceleration

Secondary electromagnetic processes

Nuclear photonics

Extreme field physics

Ultra-high intensity facilities

Pair production in a gas target as a method for the laser intensity diagnostics

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We study the process of electron-positron pair production in strong laser fields seeded by two types of gas targets: free electron gas and neutral xenon, electrons of which get ionized. Once the electrons gain sufficient energy in the presence of the laser field, they can emit high-energy photons which subsequently decay producing electron-positron pairs via the Breit-Wheeler mechanism. By detecting the resulting positrons, one can determine the value of the laser intensity by means of the one-to-one correspondences deduced in our study. We analyze two different configurations of the external field: individual focused laser pulse and a combination of two counterpropagating laser pulses. It is demonstrated that based on our computations, the laser intensity can be recovered within the range 10^{23}–10^{26} W/cm^2 with a relative uncertainty of 10–50%. It is also shown that using neutral xenon may allow one to achieve more efficient acceleration of the electrons leading to a greater positron yield (see also [1]).

The main results of our investigation are presented in [2].

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Magnetization and X-ray emission of cluster gas irradiated by polarized intense laser pulse

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Betatron radiation generated by relativistic electrons during their wiggling motion in an ion channel is a well-studied source of X-ray photons. Here, based on theory and computer simulations, we present the method to enhance X-ray power from inhomogeneous plasma irradiated by linear polarized intense laser pulse. The stronger wiggling of electrons is achieved by using cluster gas targets, which allows to get few orders of magnitude higher X-ray yield than in a uniform gas target with similar average electron density [1]. In such targets the electron trajectories are modified by strong and well localized space-charge fields of clusters bending the relativistic electrons more efficiently than the static field of an ion cavity in pure gas targets. We defined the optimum laser and cluster parameters for this process.

The analytical model and PIC simulations developed in [2,3] for the description of the interaction of cluster gas with a circularly polarized intense laser pulse were used to investigate synchrotron radiation from electrons orbiting around the cluster. The energy of radiated quantum is: $h\omega_\beta \approx h\omega\eta a^3 c\tau_L/16R$, where ω, τ_L, a – laser pulse frequency, duration and dimensionless amplitude, η – absorption coefficient and R – cluster radius. The number of quanta is: $N_\beta \approx \eta^2 a^{3/2} A^{0.5} {m_p}^{0.5} {m_e}^{0.5} \omega c^2 {\tau_L}^2/24hZ^{0.5}$, where A, Z atomic number and ion charge of the target, $m_{p,e}$ – proton and electron mass. For Au^{+30} cluster of radius 50 nm irradiated by laser pulse of intensity 10^{20} W/cm^2 and duration 8 fs one can obtain 7×10^8 quants of energy 8 keV. Conversion efficiency of emitted quanta energy in respect to laser energy can be estimated as $k \approx 10^{-2} A^{0.5} {m_p}^{0.5} \eta^3 a^{2.5} e^2 {\tau_L}^2/Z^{0.5} {m_e}^{1.5} R^3$ and for the laser and cluster parameters, mentioned above: $k \approx 10^{-2}$. The main difference of cluster synchrotron radiation is its angular distribution with a sharp maximum in transversal direction with respect to laser beam propagation where this maximum is directed along the beam in uniform plasma. More detailed comparison (taking into account particle energy distributions) of laser magneto-active cluster plasma radiation with betatron radiation from under-dense homogeneous plasma show us that at relatively low laser intensity cluster plasma generates low energy quanta and cannot compete with uniform under-dense plasma. Anyway, at laser intensity above 10^{20} W/cm^2, circularly polarized laser pulse generating cluster plasma and having optimal (about 100 nm, heavy Au) clusters, results in bigger X-ray energy in comparable (focal) volumes.

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Laser driven sources of high energy particles and radiation for multidisciplinary research

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High-current well-directed relativistic electrons is an excellent toll for applications in many research fields such as plasma physics, nuclear physics, biology, cancer therapy, material science, etc. Pilot experiments performed at PHELIX-facility in Darmstadt as well as Particle-in-Cell and Monte-Carlo simulations demonstrated strong increase of particle and photon fluence in interaction of relativistic laser pulse with long-scale plasma of near critical density. A good agreement between the experimental data and the results of the 3D-PIC simulations was obtained [1,2].

Betatron radiation of the DLA-electrons was investigated numerically for PHELIX-laser parameters (0.7 ps, 80 J, 2×10^{19} W/cm^2) [3]. The x-ray source of an ultra-high intensity was demonstrated [4], comparable to those predicted for 1 kJ PETAL-laser for Self-Modulated LWFA mechanism. High current DLA-electrons penetrating high Z convertor give rise to intense MeV bremsstrahlung radiation with record conversion efficiency of some percents, what is ten times higher than those obtained at OMEGA EP laser.

Ultra-intense well-directed beams of MeV particles and radiation were obtained at laser intensities that are relevant for the short pulse high energy diagnostic lasers for HED research, e.g. at NIF and LMJ. The use of low-density foams has paved the way to the practical implementation of DLA in NCD plasmas, requiring neither ultrahigh laser intensity nor high laser contrast.

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Laser-driven magnetic filaments as a platform for high-field science

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High-power high-intensity multi-beam laser systems that are becoming operational around the world can now be used to create a platform for high-field science that is based on relativistically transparent magnetic filaments driven by irradiating lasers within a dense plasma [1]. The strength of the quasistatic field can be comparable to that of the laser (fig. 1b), reaching the megatesla level. This talk will review several phenomena that can be studied with experimentally achievable laser intensities at multi-PW laser facilities. These include emission of dense gamma-ray beams in the quantum regime [1,2] and electron-positron pair creation from light alone [2,3]. Astrophysical environments are known for exotic physics regimes that involve generation of extreme magnetic fields and creation of matter and antimatter from light alone. The discussed platform provides a potential path towards recreating relevant regimes in laboratory conditions.

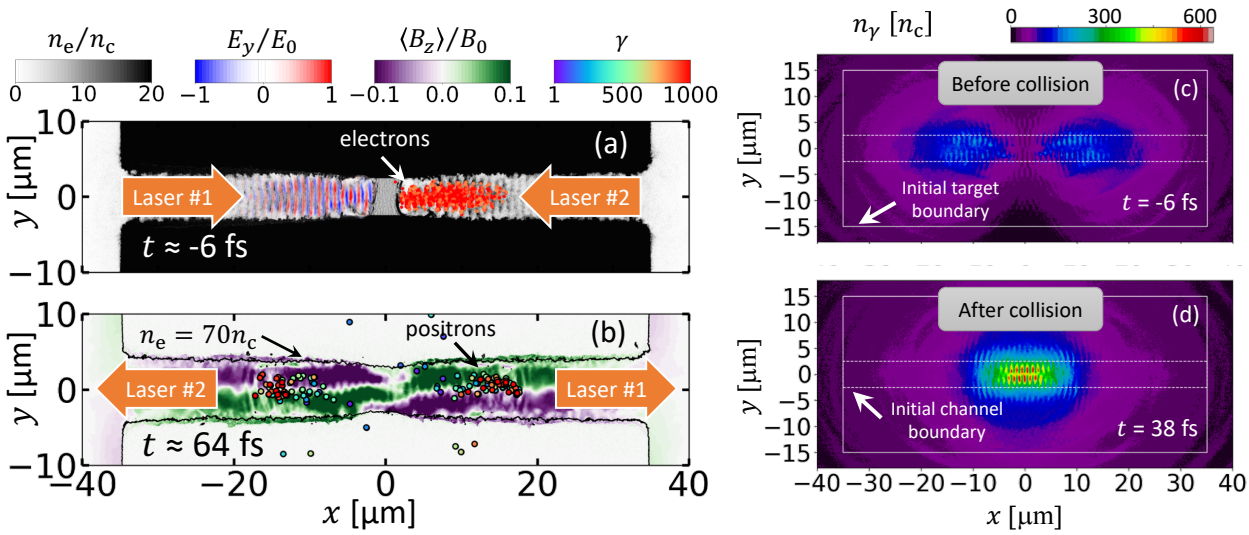

Figure 1: Structured target during irradiation by two laser ($a_0 = 190$) in a PIC simulation. (a) Electron density n_e (gray scale), electric field E_y of laser #1, and electrons with $\gamma \geq 800$ accelerated by laser #2 (dots, colored by γ). (b) Laser-accelerated positrons (points), confined by the quasistatic plasma magnetic field $\langle B_z \rangle$ (color scale). (c) and (d) Photon density ($\varepsilon_\gamma \geq 1$ keV) before and after the collision of the two lasers. E_0 and B_0 are the peak laser fields in vacuum; n_c is the classical critical density.

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QED effects and pair plasma dynamics in laser beam configurations maximizing the magnetic field

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At present modern laser facilities are on the verge of performing QED experiments and in the near future these facilities will be able to cover a wide range of fundamental problems from testing the quantum recoil effect to generating electron-positron plasmas via QED cascades and studying the properties of such plasmas under extreme conditions. Many of these quantum and plasma processes are very sensitive to the strength of electric and magnetic fields, so tight focusing of laser beams can not only decrease the threshold of their manifestation in experiments but reveal their unique properties [1, 2].

Much attention has been paid to the optimal configuration of laser beams with respect to maximizing the electric field, which plays a leading role in establishing many fundamental effects. However, the magnetic field is also a key factor in various basic physical phenomena. In particular, in such an important problem as the QED creation of electron-positron pairs by ultra-high-power lasers, both electric and magnetic fields are equally important: an electron is accelerated to high energy in an electric field, but mainly it emits gamma photons on curved paths induced by a strong magnetic field [3]. Also, gamma photons, in turn, decay into electron-positron pairs predominantly in areas of the strong magnetic field [4].

This report is devoted to a comprehensive analysis of the motion of particles, QED cascades, and the dynamics of the generated electron-positron plasma in focused laser fields that maximize the magnetic field. We will pay special attention to the case of extreme focusing corresponding to a magnetic dipole (m-dipole) wave [5]. We show that the configuration of the m-dipole field can be very promising for upcoming experiments with extreme fields, providing a unique tool for testing QED theories, creating new electron-positron plasma structures under extreme conditions, generating harmonics of incident laser radiation and gamma rays. In addition, the properties of the generated radiation and electron-positron beams make it possible to unambiguously detect extreme states of matter.

The reported study was funded by RFBR and ROSATOM according to the research project № 20-21-00095.

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Generation of transient strong shocks by fs- lasers

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The investigation of the generation of very strong shocks (initial pressures larger than 100 Mbar) [1,2] in solid density matter is important for developing a platform for diagnosing matter in extreme pressure conditions which are relevant for shock ignition conditions [3,4]. Here we report an experimental campaign, performed at the CLPU VEGA – II laser facility, focused on studying the dynamics of shock produced by fs-laser via the energy deposition from hot electrons generated in laser-matter interactions. Due to the short pulse duration, pressure is not maintained in time and the shock takes the form of a blast wave.

In the experiment, solid-density targets of various materials and thicknesses were irradiated by the fs-laser and a full characterization of the blast wave formation and propagation was performed using several diagnostics. X-ray emission was detected by using a bremsstrahlung cannon, a Kirkpatrick-Baez and GEMPix detector. Together with electron spectrometers they provided information about hot electron temperature. Optical diagnostics (shock chronometry [5]) provided information on the propagation of the blast wave in solids. Doppler reflectometry [6] provided information on the state of the target rear surface.

Preliminary experimental results confirm the generation of a blast wave which is initially very strong and very rapidly decaying in time, confirming the estimations predicated by radiation magnetohydrodynamic, hybrid Vlasov-Fokker-Planck and particle-in-cell simulations.

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Nano-metallurgy for laser thermonuclear targets and plasma research

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Metal presence in the laser thermonuclear targets has numerous purposes and varying motivation. If not to take into account the carrier- and the support- use then the needs for normal strict requirements of uniformity and mass equality on the scale of laser wavelength in the third power arize. In the microworld of laser targets the high Z metals should then be performed and treated on the nanoscale.

The topics for consideration will be:
1) the target solid and low-density layers of metals;
2) super-lattice of metal admixtures in the matrix layer;
3) equipping the targets with a low-density layer of nanoparticles on the surface;
4) all-metal solid spherical targets with thin and with thick walls;
5) alternative fuel in the layers and in laser shell targets for fusion;
6) some results with targets in the laser shot.

Thus on the real targets and on the model experiments a number of metallurgy methods are utilized and some are invented on the nanoscale. Several other techniques are combined to provide a steady and controllable realization in the form of research target and in the form of thermonuclear microtarget.

So the target design, development, measurable witnesses of the interaction targets, sample monitoring and selected research applications are proposed for your attention.

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Photonuclear applications with gamma rays from laser-accelerated electrons in a self-trapping regime

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Very efficient generation of a high-charge electron beam by a laser pulse propagating in a self-trapping mode in near-critical density plasma makes it possible to produce a high yield of gamma rays for number of practical application based on nuclear physics. Here we reports nuclear reactions of interest to shielded radiography, medical isotope production and transmutation of long-lived fission products based on Bremsstrahlung gamma beams produced from the electron accelerated in the relativistic self-trapping regime of laser pulse propagation in near-critical density plasma.

Laser pulse with a peak power of several hundred TW and a pulse duration of tens of fs propagates in relatively dense plasma as a "laser bullet" that is a trapped light in a plasma cavity, i.e. electromagnetic-plasma structure of a soliton type [1]. As long as the laser pulse propagates (with almost speed of light) to its depletion, it accelerates electrons to energies of tens to hundreds of MeV. The accelerated electrons leave a plasma and are sent to the converter to produce Bremsstrahlung gammas [2]. We illustrate the capabilities of the proposed scheme for single-shot gamma radiography of samples located deep in a dense medium [3] by using a compact laser system. We also evaluate the efficiency of photonuclear transmutation of a number of long-lived isotopes and photonuclear production of a number of medical isotopes for positron emission tomography (PET) and single-photon emission computed tomography (SPECT).

This work was supported by by the Russian Science Foundation (Grant No. 17-12-01283).

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Orbital angular momentum absorption by an electron in a laser wave

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Generation of quasistatic magnetic field is observed in numerical simulations of the interaction of spatially structured laser beams carrying orbital angular momentum (OAM) with a dilute plasma. Magnetic field generation is caused by a rotation of electrons, that have absorbed a fraction of the electromagnetic wave OAM[1]. Numerical simulations and the detailed analysis of the process show that the gain of angular momentum may be considered as a single particle effect.

Analytical consideration of the process may be simplified by using the paraxial approximation. It is widely used to prescribe propagating electromagnetic fields with moderate focusing. In general case symmetries present in an infinite plane wave are destroyed in paraxial approximation, however configurations of the fields, preserving some of the symmetries still exist. These symmetries manifest themselves in emergence of the integrals of motion, that may be used to characterize properties of the interaction. In a low intensity regime, the paraxial approximation should be reconsidered for correct description of the orbital angular momentum absorption.

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On generation of ultrashort pulses X-ray pulses by the inverse Compton scattering

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Ultrashort X-ray pulses with duration in the picosecond range may find various applications in diverse areas of condensed matter physics, material science, chemistry and molecular biology [1]. These applications may include, for instance, observing phase transitions in solids in real time as well as X-ray structural analysis of transient states including lattice deformations caused by laser heating. Other applications include studying fast processes such as heat and phonon propagation in solids, chemical reactions processes in solutions, solid-state decomposition, and explosion of energetic materials as well as studying evolution of the structure of complex proteins with picosecond temporal resolution. Picosecond X-ray pulses can also be used to study phase transitions in chalcogenide alloys such as $Ge_2Sb_2Te_5$ (GST) or similar materials, which find applications in Phase Change Memory (PCM) vying as a replacement of DRAM and FLASH memories [2].

While there are many ways of generating ultrashort x-ray pulses, which are based on laser plasmas, synchrotron radiation, x-ray lasers and X-ray tubes, the existing methods do not satisfy the needs of all users. Therefore, in this presentation the design, X-ray radiation parameters and possible applications of the Picosecond Compton X-ray Sources (PCXSs), which are based on scattering of short and energetic laser pulses by trains of low emittance electron bunches produced by a linear accelerator, are considered. It is demonstrated that creation of such sources is feasible on the basis of existing accelerator and laser technologies. The design of both the linear accelerator and the main laser system of PCXS are discussed as well as possible design of the X-ray optical system delivering X-ray pulses onto the sample is also considered.

Creation of such a source of ultrashort X-ray pulses can significantly advance the physics of fast processes in such fields as studying the fast structure changes of complex proteins, the evolution of transient states in solids and the dynamics of fast chemical reactions.

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X-ray radiography measurements of short-lived hydrodynamic phenomena in astrophysically relevant plasma flows

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The study of a laser plasma with large gradients of density is of a great interest for many branches of science, including laboratory astrophysics [1,2], warm dense matter (WDM) [3] and many others. Typically, the measuring of plasma parameters and dynamics is quite challenging due to short temporal and spatial scales. By the way, diagnosed objects are often opaque to the probe beam in a visible range of wavelengths. For the tasks of laboratory astrophysics, an experimental high-quality pulsed radiographic research of short-lived plasma was developed using the interaction of intense laser with solid matter. Pulsed x-ray radiography allowed us the study of fast evolving phenomena like shock compression of matter or plasma outflows, interactions with gas or plasma atmosphere reducing smearing of even high velocity phenomena ($v > 100$ km·s^{-1}) to the µm scale. Using a thin 20 µm wire, reproducible high quality x-ray radiographic images in a new vertical configuration of x-ray source and detector were demonstrated successfully while the laser irradiated on the target in the horizontal plane. Questions related with a finding of optimum between such parametersas a spatial resolution, contrast, generation of hot electrons which are responsible for the deterioration of the image quality, are considered. Spectral composition of the backlighter radiation was recovered by means of the special filtering before the detector. As a result, an example on a laboratory astrophysics demonstrated a high resolution and a linear density map of the plasma obtained with this configuration in the experiment simulating cataclysmic variables in a Polar experiment. In the experiment laser pulses with 1.5 ns, 600 J were focused to 300 µm focal spot at solid laminate target in presence of 20 T poloidal magnetic field.

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Locally constant crossed field approximation for the radiation in a time-dependent electric field

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Despite the tremendous progress of the laser technology in the last 25 years the high intensity laser systems are not yet generally available, and therefore numerical simulations are widely used for a study of high field phenomena, e.g. radiation friction, gamma generation, QED cascades, etc. The exact QED rates for the basic processes – photon emission and electron-positron pair creation are known only for the simplest configurations such as a constant electromagnetic field or a monochromatic plane wave. Therefore approximations should be used for implementation of QED processes into the numerical codes.

Most of the modern codes employ Locally constant crossed field approximation (LCFA) [1], assuming that

- particles in a strong field are ultrarelativistic, and therefore the field is crossed in the proper reference frame;
- the formation length of the processes is much shorter than the field wavelength, and therefore the field can be considered as locally constant.

LCFA is strictly justified for a plane wave like [1,2] and magnetic fields [3]. However, the list of possibly interesting field configurations is much wider. In particular, colliding laser pulses, producing standing wave like field, are more preferable for QED cascades generation than a single pulse [4–6]. In the electric field antinode of the standing wave the magnetic field vanishes, and the time-dependent electric field can serve as a model providing quantitative estimations [5].

We develop the approach for the calculation of the photon emission probability in a generic time-dependent electric field. We derive LCFA and establish the conditions of its applicability. Our approach allows to calculate any order corrections to LCFA and we provide ones of the first and second orders.

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Analytic model for excitation of a plasma wakefield in the bubble regime

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Plasma-based acceleration methods are promising for achieving extremely high energies of accelerated particles at short distances [1]. Their core principle is to use a plasma wakefield excited by a laser or particle driver for acceleration. When the laser intensity or particle bunch density is sufficiently high, interaction with plasma happens in the strongly nonlinear regime. In this case, an almost spherical cavity devoid of plasma electrons (referred to as a "bubble") is formed behind a driver. This regime provides both high acceleration gradients and focusing of the accelerated electrons.

Theoretical description of the bubble regime proves to be difficult due to its highly nonlinear nature. A major breakthrough was achieved when a phenomenological model of the bubble was created and an equation for its boundary was derived [2]. This model is capable of predicting the shape of the bubble and the distribution of electromagnetic field within it, taking into account beam loading effects. However, a major downside of this model is that it cannot self-consistently describe the properties of the wakefield based only on the properties of the driver.

In this work, we present a new analytical model for the description of the bubble excitation by an electron driver [3]. The model is based on using the energy conservation laws and solving the equation for the bubble's boundary in the relativistic approximation starting from the axis of the bubble at the front of the driver. To circumvent the singularity in the equation, a substitution is suggested, making the numerical solution possible. The validity of the model is verified with 3D particle-in-cell simulations. For a special case of a cylindrical driver, a fully analytical solution is derived. Based on the analytical solution, scaling laws for the properties of the bubble depending on plasma and driver parameters are presented. We also discuss the possibility to generalize this model for laser drivers.

The work was supported by RFBR (projects № 20-52-12046, 20-02-00691).

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THz radiation generation in the interaction of TW laser pulse with dense tens of micron-scale preplasma layer

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The field of research devoted to the generation of THz radiation in relativistic laser-plasma interaction is currently under active investigation [1]. However, the generation of terahertz radiation due to the interaction with a highly inhomogeneous plasma ($L \sim \lambda$) is hardly studied. There is still little discussion about the role of parametric processes in the process of THz radiation generation, and such discussions are usually focused on subrelativistic intensities. It's also worth noting that the source of the prepulse is usually the amplified spontaneous emission (ASE) of the laser itself and therefore cannot be controlled. Thus a more detailed investigation of THz radiation generation in intense laser-matter interaction should take place.

In our recent paper an efficient injection and acceleration of electrons using relativistic laser pulse with normalized vector potential $a_0 \sim 2$ was demonstrated experimentally in plasma layer with density $\sim 0.3n_c$ and thickness of the order of tens of microns [2]. Acceleration mechanism was found to be is Direct Laser Acceleration (DLA) in the plasma channel formed by the laser pulse. Injection mechanism was found to be breaking of plasma waves of parametric instabilities.

Experiments were carried out at the 1 TW Ti:Sa laser facility in Lomonosov MSU (800 nm, 50 mJ, 50 fs, 10 Hz). The peak vacuum intensity was 5×10^{18} W/cm^2. Laser polarization cold be changed from linear p-polarization to elliptical by adding a PET pellicle. We used 16 µm thick audiotape as a target. An additional Nd:YAG laser pulse (1064 nm, 10 Hz, 200 mJ, 10 ns) was used to preform the target in a desired manner. Prepulse intensity was $\sim 5 \times 10^{12}$ W/cm^2. We experimentally observed electron beam with divergency of ~ 0.05 rad, charge of ~ 50 pC for particles with $E > 1.7$ MeV with the pulse energy as low as 30–50 mJ.

No studies of THz radiation generation with similar experimental parameters had taken place earlier. We used a Golay cell to detect THz radiation in the frequency range < 10 THz. A characteristic feature of obtained THz radiation was found to be its two-maxima structure arising with the change in preplasma electron density. THz radiation spectra was also measured and is located in 1–3 THz range. Possible mechanisms of THz radiation generation will also be presented, as well as other discovered features.

This work was supported by RSCF grant 20-79-00051. D.A.Gorlova acknowledges Foundation for the advancement of theoretical physics "BASIS" for the financial support.

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Laser triggered stochastic electron acceleration in dusty plasma irradiated by a powerful pulse of record short duration

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Modern technologies make it possible to shorten the duration of PW laser pulses to about 10 fs. At the same time, there is an active worldwide search for various mechanisms of particle acceleration and interaction schemes for a laser target in order to increase the yield and energies of accelerated electrons and ions. The nano- and micro-structured targets, including droplets and micro clusters, play an important role in this context. The absorption of laser energy in cluster plasmas is efficient due to good transparency of cluster gas and high average density of particles.

In our work the matching condition for laser-cluster parameters is established, which makes it possible to maximize the yield of hot electrons with required energies upon irradiation of an ensemble of submicron-sized clusters with an ultrashort laser pulse. Using 3D PIC simulations, we studied the interaction of relativistic femtosecond laser pulse with submicron-sized cluster plasma. The propagation of a laser pulse through such targets leads to forming a plateau in the energy spectrum of electrons, which is important for applications. For a given laser energy, which depends on the radiation intensity, which changed simultaneously with the focal spot diameter, the dynamics of both groups of energetic electrons stochastically wandering between clusters and recirculating around them were studied. The important role of the stochastic acceleration mechanism in the Coulomb fields [1] for the generation of a large number of hot electrons with energies of the order of the ponderomotive one is established. It has been shown that the yield of hot electrons reaches a value of 6×10^{12} particles per 1 J (or in terms of a charge of 1 µC per 1 J) for electrons with energies above 100 keV, and for electrons with energies above 300 keV output is 0.5 µC per 1 J.

This work was supported by RFBR and ROSATOM, grant №20-21-00023.

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High-Z structured target for efficient laser-driven X-ray source and phase-contrast imaging applications

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The parameters of relativistic laser-driven plasma X-ray source, formed on the surface structured high-Z (copper and tungsten) target were investigated. It was demonstrated that structured target provides fewfold growth of photons flux compared to that from the flat substrate up to the peak intensity 2×10^{18} W/cm^2 (the ASE contrast level of 10^{-9} a hundred of picoseconds in front of the main pulse). This limitation is caused by the lowered ablation threshold of the structured target (the estimated value is ~ 0.2 J/cm^2) and explosion of the structures under the irradiation by ASE pedestal prepulse with the use of the main pulse with higher peak intensity. We detected a maximum conversion efficiency into the linear radiation around 1.4×10^{-4} at the peak intensity $\sim 1.5 \times 10^{18}$ W/cm^2 for a laser pulse with energy ~ 20 mJ on target. Though the absolute yield of K_α photons grows continuously with peak intensity increase and reaches 4×10^9 photons at maximal intensity 2.5×10^{18} W/cm^2. The approach of using a structured target is promising for X-ray laser-driven source optimization with high contrast ($\geq 10^{-10}$) laser pulses at peak intensities up to 10^{19} W/cm^2, when the structures remain unaffected up to the time of the main pulse arrival providing enhanced absorption and fast electron production.

The measurements of X-ray source size revealed that the spot diameter fluctuates near 15 µm for the flat and structured targets, making both suitable for the X-ray phase contrast imaging using a reasonable scheme allowing to maintain high flux on the detector for the fast image accumulation. At the same time X-ray source shot-to-shot shifts hampers the acquisition of high-resolution images. We showed the simple method to obtain images in each laser shot with a marker in order to track the displacement of the source. Using an algorithm to shift each frame before summing one could retrieve the lateral coherence corresponding to net source size and detect the phase contrast effects near the border of the object in the case, when the source shot-to-shots shifts exceeded its diameter. The estimated contrast C was 5–10% with the spatial resolution of ~ 15 µm. With a proper marker alignment along the camera pixels the image quality may be further increased. The proposed approach is useful when multi-shot exposing is needed.

The work was supported by RSF grants 18-79-10160 (structured target manufacturing) and 21-79-10207 (X-ray detectors).

Optically self-photopumped X-ray laser at ~ 13.4 nm in Sn^{22+} in plasma produced by intense pump laser interacting with nanostructured Sn target, as promising radiation source for nanolithography.

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Scientific and technological progress of recent years predetermines new directions for the next-generation nanolithography, i.e., production and control of semiconductor chips < 20 nm in size, based on three principal developments:

(i) Development of multilayer mirrors (MMs): for Mo/Si MMs at $\lambda = 13.4$ nm, the reflectivity can be increased to 70% [1].

(ii) Development high-efficiency X-ray lasers (XRL) for these wavelengths in nanostructured targets with longitudinal optical pumping. The plasma formation mechanism is ionization by a strong optical field. Due to the extremely narrow XRL linewidth $\sim \Delta\lambda/\lambda \leq 10^{-4}$, and high degree of coherence XRLs can be ideal sources for commercial nanolithography (interferometry, microscopy, and high-resolution holography).

(iii) Development of high average power, diode pumped Petawatt laser systems: a new generation of lasers enabling precision science and commercial applications [2]. Under certain conditions, these lasers can be used for plasma pumping for generating XRL radiation. The high conversion efficiency of the pump energy to the XRL energy is achieved when using cluster/nanostructured targets.

A new class of the self photo-pumped X-ray lasers (SPPXRL) was studied experimentally in Ref. [3] on the $3d^9{}_{3/2}4f_{5/2}[J = 1] - 3d^9{}_{3/2}4d_{5/2}[J = 1]$ transition of Ni-like ions Zr^{12+}, Nb^{13+}, Mo^{14+}. Solid targets for these materials were used in [3]. The list of elements that lase on the self-photopumped transition can be extended much further than originally known. We have calculated the wavelengths of this transition in Ni-like sequence to Z = 79 using the relativistic perturbation theory with a zero-approximation model potential (RPTMP) [4]. For Z = 50 (Ni-like tin), $\lambda_{las} = 13.41$ nm. The gain of the X-ray laser with $\lambda = 13.4$ nm in Sn^{22+} is calculated under the assumption that plasma is formed during the interaction of a nanostructured (cluster-like) tin target with high-intensity pump laser radiation. The duration of the outgoing X-ray laser pulse does not exceed 10-20 ps. Thus, special equipment for the production of nanostructured targets and the formation of plasma column must be developed. The optimum plasma density, temperature, geometry, and pump parameters are determined to achieve the highest Sn^{22+} ion fraction and emission quantum yield. The accuracy of our calculation was checked by comparing of our calculation of the SPPXRL parameters with known experimental data for Ni-like molybdenum (Mo^{14+}).

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Boosting the efficiency of high harmonic amplification by modulating a plasma-based X-ray laser with the second harmonic of the fundamental frequency laser field

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Attosecond pulses formed by high order harmonics (HHs) of an infrared (IR) laser field is a powerful tool for studying and controlling ultrafast dynamics of electrons in atoms, molecules and solids at its intrinsic time-scale. However, in the X-ray range the energy of attosecond pulses is rather limited. Their amplification is an important but very challenging problem since none of the existing amplifiers can support the corresponding PHz bandwidth.

In our previous work [1] we proposed a method for the attosecond pulse amplification in hydrogen-like active medium of a recombination plasma-based X-ray laser dressed by a replica of the fundamental frequency IR field used for the HH generation. Due to the IR-field-induced sub-laser-cycle Stark shift and splitting of the lasing energy levels the gain of the active medium is redistributed over the combination frequencies, separated from the resonance by even multiples of the frequency of the IR field. If the incident HHs forming an attosecond pulse train are tuned in resonance with the induced gain lines and the active plasma medium is strongly dispersive for the modulating IR field, then during the amplification the relative phases of harmonics and (under the optimal choice of the IR field strength) the shape of the amplified pulses will be preserved.

In the present work we show the possibility of boosting the efficiency of HH amplification by modulating the active medium of an X-ray laser with the second harmonic of the fundamental frequency IR field. We show that under the action of a laser field (with arbitrary frequency) the gain redistribution occurs not only over the even combination frequencies discussed in [1], but also over the odd frequencies separated from the resonance by odd multiples of the laser frequency. Besides, nearly half of the medium gain is contained in the even induced gain lines, and nearly half in the odd. If the modulating field is the second harmonic of the IR field, used for the generation the HHs and attosecond pulses, then the seeding HHs can be tuned in resonance with both even and odd gain lines simultaneously, which will make the overall gain much higher as compared to the previously considered case of the fundamental frequency modulating field (when only the even gain lines play the role). By the example of the C^{5+} X-ray laser with 3.38 nm wavelength of the inverted transition we show the possibility of increasing the efficiency of 430 as pulse amplification by 8.5 times when the active medium is modulated with the second harmonic of the fundamental frequency IR field with wavelength 2.1 µm.

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11 fs, 1.5 PW laser with compression after compressor approach

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Compression AFter Compressor Approach (CafCA) were used to compress the 60–70 fs laser pulse to 11 fs after passing through a 5-mm thick silica plate and reflecting from two chirping mirrors with a total dispersion of –250 fs^2. The experiments were carried out for the B-integral values up to 19 without damage of the optical elements, which indicates that small-scale self-focusing was suppressed. To the best of our knowledge, the duration of 11 fs is the shortest for all petawatt lasers worldwide. Taking into account the obtained results and undoubted merits of nonlinear compression (simplicity, low cost, negligible pulse energy losses, and applicability to any high-power laser), we predict further development of this approach toward multi-PW power and single cycle pulse duration simultaneously.

The first Rytov approximation and its limits in describing the wave diffraction behind a micrometer-sized phase object

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The diffraction spreading of a plane electromagnetic wave behind an axisymmetric phase object with a micron size is simulated. Modeling is carried out by employing the first Rytov approximation. The convergence of the Rytov approximation is investigated depending on the step of the computational grid in the transverse and longitudinal directions of the wave propagation. The computational code is verified by simulating the wave diffraction by different test objects having different distributions of the refractive index. It is shown that there is a discrepancy between the simulation results and the results obtained by calculating the spreading of the angular spectrum of the diffracted wave when solving the homogeneous Helmholtz equation. The zone behind the phase object with a reliable coincidence of the simulation results obtained by the two independent methods is determined. The distributions of the characteristic errors of the simulation results are constructed for the region behind the phase object. The limits of the employment of the first Rytov approximation in describing the diffraction spreading of an electromagnetic wave in free space are discussed.

The study was supported by the Russian Science Foundation (grant no. 19-79-30086). Theoretical analysis and data processing were funded by the grants of the Russian Foundation for Basic Research (no. 20-08-01156) and the President of the Russian Federation (no. MK-703.2020.2).

Transient processes in micro-coils induced by a short laser pulse

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When a short laser pulse irradiates a target with a considerable spatial scale, transient processes become important under the condition $\tau \ll L/c$, where τ is the pulse duration, L is the target spatial scale, and c is the light velocity. In this case, a transient discharge pulse is excited and propagates along the target surface with the velocity close to the light velocity. For laser pulses of ps duration, the corresponding target scale L should be greater than several hundred microns, while for fs laser pulses, transient processes play an important role already for micron-size targets. For intense laser drivers and therefore intense discharge pulses, they propagate in a nonlinear regime, forming behind hot expanding plasma carrying magnetic fields. In this case, energy in the discharge pulse gradually decreases and the dispersion may noticeably reduce its group velocity. Using a coil-bent wire as a target allows to create in this regime quasi-stationary magnetized structures, living for hundreds of ps after a very fast interaction stage. Some schemes, involving additional geometrical constraints of the interaction, like a grazing incidence along a curved cylindrical surface, may rich an efficiency of $\sim 10\%$ for conversion of laser light to magnetic field.

Within certain conditions, the discharge pulse may be visible while propagating for several hundreds of microns. Providing a small dissipation, it may be reflected, divided on several parts and pass along the same surface multiple times. With numerical simulations, this regime is studied for different parameters of interaction in relativistic intensity domain for micro-coil targets. Possible applications of the observed phenomena are considered.

Simulation of generation of characteristic X-ray radiation under vacuum heating of electrons of nanocylinders

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Investigation of the K_α radiation from nanowire targets may advance development of efficient and compact sources of narrow band X-ray emission driven by moderate-intensity lasers.

We have previously developed a model of the generation of characteristic X radiation under vacuum (Brunel) heating of electrons by a femtosecond laser pulse of nonrelativistic intensity [1]. This model satisfactorily describes measured K_α yield from massive iron target depending on the laser pulse energy, and measured enhancement of K_α yield from massive silicon target covered by closely packed spherical clusters, as well as the average electron energy, depending on the cluster radius, multiplied by the laser wavenumber.

In this paper, we consider a model of generation of K_α radiation in a copper foil under vacuum heating of electrons of ionized nanocylinders deposited on the foil obliquely and parallel to each other. A laser pulse with the normalized electric field amplitude equal to 0.2 is directed perpendicularly to the foil. Dependences of the K_α yield on the cylinder radius, multiplied by the laser wavenumber, and on the angle of inclination of the cylinders, as well as on the angle between the polarization vector and the plane, formed by the normal to the foil and the axis of a cylinder, are obtained. The maximum yield of K_α radiation from the foil covered with nanocylinders exceeds the maximum yield of K_α radiation from the foil covered with clusters by about three times. The K_α yield depends rather strongly on the angle of inclination of the cylinders, while it changes little with a change of the polarization angle in a wide range. The radiation yield weakly depends on the permittivity of the nanocylinders. The characteristic geometric parameters of the target are determined.

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Self-trapping of extreme radiation

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A modern understanding of the self-focusing regime of a laser pulse self-trapping in a strongly nonlinear medium predicted almost 60 years ago is presented on the example of a relativistic plasma. A theoretically substantiated proof of the possibility of its implementation is given both with the help of simple qualitative and simplified model approaches, and with the help of an analytical approach based on the nonlinear Schrödinger equation (NSE). An analytical approach to the NSE, based on exact solutions to the problem of self-trapping in plane geometry, makes it possible to qualitatively substantiate and predict the results of numerical simulation of a self-trapping mode. An approximate analytical approach to the NSE in cylindrical geometry [1], which is as close as possible to a real experiment, enables to predict the main regimes of the propagation of a relativistic beam in various modes, including self-trapping. For the latter, this approach made it possible to obtain a physically and theoretically substantiated condition for matching the laser spot size with the plasma density and laser pulse intensity, which is in good agreement with the results of multidimensional numerical simulation.

This work was supported by the Russian Science Foundation grant № 17-12-01283.

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The Markoff approximation for high harmonic generation at the laser–atom interaction

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The purpose of this talk is to evaluate the high harmonic power spectrum of the spontaneous radiation irradiated by an atomic system driven by a strong laser pulse. The scattered radiation possesses, in addition to a monochromatic, elastic component, inelastic components that are the result of the alteration of the atomic continuum states by the time-dependent laser field. As the intensity of the driving field is increased, inelastic scattering begins to contribute, becoming appreciable when the multiphoton matrix element V becomes comparable to the natural relaxation rate γ. In the limit of strong driving fields, inelastic scattering predominates over elastic scattering. The N-harmonic power spectrum in this limit has peaks centered at $\nu = N\omega$ (the incident laser field frequency is ω) and at the displaced frequencies $\nu = \omega \pm 2V$, with widths proportional to the atomic relaxation rate γ. The effect of radiation damping emerges naturally as the result of the coupling of the atom to the quantized electromagnetic field modes into which the atom radiates. In our previous paper [1], we developed a quantum theory describing the spectrum of photons emitted by a generic nonstationary atomic system and showed how to implement it in practical calculation. To illustrate the theory, we applied it to calculating high-order harmonic generation spectrum for a one-dimensional zero-range-potential model. The applicability of the treatment of ref. [1] requires $\gamma T \ll 1$ where T is the duration of laser pulse. This condition ensures that radiative processes do not affect the electronic motion. In this paper we consider the opposite limit $\gamma T \gg 1$ when stationary states occur. Generally, the operator for the total density matrix of the atomic system irradiated by laser field must also depend on the operator for the creation and annihilation of photons of the vacuum electromagnetic field. However, if it is assumed that the changes that occur in the atom have little effect on the vacuum states, then the operator for the total density matrix can be expressed as a product of two operators; one that depends on the operators for the atom+laser, and the other that depends on the operators for the vacuum photons. This is what is meant by the "Markoff" approximation. However, it cannot be applied when $\gamma \propto N\omega$. In such a case the time required for the emission of a photon, $1/\gamma$, is comparable to the time it takes the atom to change from one state to the other,. This follows from the time energy uncertainty principle. If the operator for the total density matrix is averaged over the vacuum photon states according to the Markoff approximation, one obtains the operator for the atomic density matrix ρ. This operator enables one to predict changes in atomic states without having to analyze the states of the vacuum electromagnetic field. In the linearly polarized laser field an ejected electron after the tunneling ionization oscillates: it can return back and scatters on the atomic core approximately in half of a field period. During the classical motion this electron obtains some energy from the laser field which is of the order of ponderomotive energy. In the strong laser field this energy is much higher than the ionization potential. The returning electron can recombine into the initial bound atomic state $|0\rangle$. The excess energy is taken by a spontaneously emitted photon with energy nearly of $N\omega$, $N \gg 1$. We investigate here only this third stage of the three-step model. The maximum photon energy is $E_{max} = N_{max}\omega = 3.173U_p$. Therefore, the continuum state $|1\rangle$ in our approach can be varied near this value. Correspondingly, in order to observe triplet of the photon power spectrum, in experiments it is needed to separate narrow intervals of photons frequencies. The width of this interval should be less than V. Details of this talk can be found in our recent paper [2].

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Efficiency for terahertz pulse generation using a complex nanostructured target

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Terahertz pulse generation in a super-intense laser radiation interaction with a complex nanostructured target consisting of nanowires or nanofoil stripes is studied. During propagation of a relativistic laser pulse along a nanowire (longitudinal interaction), dense high-charge bunches of electrons are forced out of the target and accelerated in the laser field, generating high-power electromagnetic radiation with various spectral composition including terahertz and infrared ranges [1]. For a relatively long laser pulse (tens or hundreds of femtoseconds) with a smooth shape, nanowire electrons are displaced from the target by each laser half cycle. In this case, the low-frequency part of the generated radiation can have a unipolar shape with characteristics determined by the laser and target parameters. Such a shape of radiation allows to add coherently emission from different target nanowires inside the laser spot. Due to nanometer scale transverse dimensions of the nanowire, laser field amplitude decreases only in its vicinity after interaction. Then a complex target can be formed by parallel nanowires each using its own part of the laser front. Also, for a relatively long laser pulse, its amplitude decreases when interacting with the nanowire only for the first few periods. In this case, consecutive interaction of a laser pulse with nanowires can be used, and a complex target can be formed from the consecutive nanowires. Moreover, using several regularly located nanowires (or nanofoil stripes), one can engineer the shape of the generated radiation pulse or even produce a train of terahertz or infrared pulses with controlled delay between them.

Efficiency for generating low-frequency radiation during laser pulse propagation along a nanowire can be significantly higher than in other geometries of interaction. In longitudinal interaction, the displaced electrons and the laser pulse propagate in the same direction for some time, which increases the duration of their interaction, while the field amplitude decreases weakly because of a small cross section of a nanowire. Besides, using multiple parallel nanowires in the complex target irradiated by a single laser pulse or using the laser pulse repeatedly until its full depletion through interaction with consecutive nanowires in the complex target increase the efficiency even more. So the main advantages of using complex nanowire targets are the possibility to engineer the shape of the radiation pulse and increasing the efficiency for terahertz pulse generation.

Using numerical 2D simulation, characteristics of infrared and terahertz radiation pulses are found for complex nanowire targets. Conversion efficiency of the laser pulse energy into the energy of generated radiation is estimated. It is shown that the conversion efficiency of the complex target can be several times higher than those for the single nanowire. This work was partially supported by the Russian Foundation for Basic Research project 19-52-45035-Ind-a.

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Optimization of a laser plasma based X-ray source according to warm dense matter absorption spectroscopy requirements

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X-ray absorption spectroscopy (XAS) [1] diagnostic has been proved to be an effective tool for warm dense matter (WDM) experimental studies. However, XAS requires a short-lived X-ray source (XRS) of sufficiently high emissivity and absence of intense characteristic lines in a spectral range of interest. In our resent study [2], we discussed choosing its optimum material and thickness to get a bright source in the wavelength range of 2–6 Å (∼2-6 keV) considering relatively low-Z elements. We demonstrated that the so-called photorecombination region of X-ray characteristic spectral emission is best suited for XAR using a laser-generated X-ray source, due to its featureless spectra of high intensity. Performed experiments showed that the highest emissivity of solid aluminium and silicon foil targets irradiated with a 1 ps high-contrast sub-kJ laser pulse of Vulcan PW laser facility is achieved when the target thickness is close to 10 µm. An outer plastic layer increases the emissivity even further [3].

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Nonlinear absorption of laser radiation due to relativistic plasma resonance in an inhomogeneous plasma

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An analytical theory of the nonlinear resonant absorption of electromagnetic radiation in an inhomogeneous plasma due to relativistic plasma resonance is developed for the first time [1]. Taking into account the relativistic nonlinearity of the excited plasma field in the vicinity of the critical plasma density [2] significantly advances the boundary of the theoretical description of resonant absorption by a laser plasma into the region of high laser intensities. The coefficient of nonlinear resonance absorption as a function of four laser-plasma control parameters – laser intensity, the scale of plasma inhomogeneity, its temperature and the angle of incidence of laser radiation on the plasma is obtained. The nonlinear effect of the plasma resonance field amplitude suppression and "switching off" of resonance absorption with increasing laser intensity is demonstrated, which is a consequence of the relativistic nonlinearity of the electron plasma in the critical density region. It is shown that the suppression effect manifests itself most significantly in the case of a large plasma corona, at a plasma inhomogeneity scale $L \geq 100\lambda$ (λ – laser wavelength), whereas at a sufficiently steep plasma inhomogeneity, $L < 10\lambda$, the effect is not so pronounced. This is illustrated in terms of the field amplitude renormalization in the region of the critical plasma density, which arises in the presented theory of relativistic plasma resonance. In turn, the renormalization leads to a redefinition of the region boundary of relativistic hydrodynamic model applicability (in terms of the laser pump intensity) [3] due to the fact that the plasma wavebreaking occurs at higher laser intensities.

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Comparative study of efficiency of ion acceleration from nanoscale structured targets at relativistic laser-plasma interaction

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The experimental results of ion acceleration on the relativistic laser-plasma interactions of femtosecond laser pulse with micro- and nanoscale structured and flat targets with ultrahigh (with XPW contrast level $< 10^{-10}$) contrast level are presented. In our experiments we used an ultrafast pulse from the Ti:Sa laser system (pulse duration — 50 fs, energy on target — up to 30 mJ, wavelength — 800 nm, repetition rate — 10 Hz, peak intensity — up to 5×10^{18} W/cm^2).

Ions were registered with the use time-of-flight magnet spectrometer with the charge-to-mass (Z/M) separation and Thomson parabola mass spectrometer.

Direct comparative experiments were carried out on the acceleration of ions on the surface of structured targets: it was demonstrated significant increase of high-Z and decrease low-Z ion flux on nanostructured target compared to flat bulk target. Also the maximum energies and temperatures was compared for different scale of structured targets. It is shown that ions are accelerated not by the electron component with the highest temperature, but by the component with the largest number of electrons. [1]

Experiments were supported by Russian Fund for Basic Research grants № 19-02-00104 and 19-02-00740.

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Particle dynamics governed by radiation losses in extreme-field current sheets

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Particles moving in current sheets under extreme conditions, such as those in the vicinity of pulsars or those predicted on upcoming multipetawatt laser facilities, may be subject to significant radiation losses. Earlier works show that radiative effects can significantly change individual, as well as collective, particle dynamics [1-6]. In this paper we investigate theoretically and numerically the influence of radiation losses on dynamics of ultrarelativistic particles in model extreme current sheets.

In the Landau-Lifshitz radiation reaction force model, when quantum effects are negligible, an analytical solution for particle trajectories is derived. In order to verify our solution and determine the range of its applicability, we solve equations of motion numerically with radiation losses within different approaches, including the semiclassical approach as the benchmark.

The applicability region of the obtained solution is determined and analytical trajectories are found to be in good agreement with those of numerical simulations with account for radiative effects. Based on these results we gain new insights into current sheet phenomena expected on upcoming laser facilities.

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Optical registration of fine-structured electrical sparks

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Formation of fine-structured electrical sparks generated in laboratory gas discharges is an intriguing phenomenon highly relevant in terms of fundamental and applied physics. The existence of the spark fine structure has recently been unveiled by laser probing techniques, implemented in high-performance optical registration systems. At the same time, fine-structured sparks naturally formed in electrical gas discharges are challenging objects of optical research. The veracity of the spark structure image obtained by laser probing techniques is still a subject for discussion due to possible distortions introduced by the employed optical setup. Herein, we thoroughly analyze this issue by simulating the spark image formation and evaluating the effect of the setup response function on the spark pattern quality. The latter turns out to dramatically suffer from the defocusing effect, whereas the spark fine structure is reliably resolved only by optics having a spatial resolution close to several micrometers.

The study was supported by the Russian Science Foundation (grant no. 19-79-30086). Theoretical analysis and data processing were funded by the grants of the Russian Foundation for Basic Research (no. 20-08-01156) and the President of the Russian Federation (no. MK-703.2020.2).

Effect of e^+e^- pair production on generation of magnetic field driven by radiation reaction

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Generation of the axial quasi-stationary magnetic field in the interaction of extemely intensive circularly-polarized laser pulse with thick plasma target is considered. When a dissipation in form of radiation reaction is present, angular momentum carried by the circularly-polarized laser pulse can be absorbed by the electrons of plasma [1], giving rise to long-living azimuthal current and thus axial magnetic field of the order of magnitude up to several Giga-Gauss when PW lasers are considered [2]. Recently it has been shown [3, 4] that when the intensity of the laser radiation exceeds 10^{24} W/cm^2 the magnitude of the generated magnetic field saturates due to the decrease of the power of the synchrotron radiation in the quantum regime compared to the classical one.

In this work this process is investigated with full 3D PIC simulation taking into account quantum-electrodynamical (QED) processes. It is shown that when decay of the gamma-quanta emitted by the plasma electrons into the electron-positron pairs is accounted for, the process of the quasi-stationary magnetic field generation changes qualitatively. In particular, when pair production is present but scarce, generation of the magnetic field is suppressed compared to the case when pair production is not accounted. At greater intensities a dense electron-positron plasma is formed as a result of QED cascading inside the cavity formed in the target by the intense laser pulse. Opposite to the previous case, formation of pair plasma increases efficiency of the magnetic field generation. Overall, this leads to the non-monotonous dependency of the amplitude of the magnetic field from the laser intensity which is qualitatively different from the monotonous saturating dependency when pair production is not accounted for. Qualitative description of this effect is discussed.

The work is sponsored by Russian Science Foundation (Grant No. 20-12-00077).

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X-ray, gamma and relativistic electron sources with tabletop femtosecond laser

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We present an overview of our recent activity on interaction of relativistic femtosecond laser pulses with dense plasma. Preplasma tailoring allows to achieve efficient generation of X-rays, gammas and well collimated electron beams with the tabletop terawatt femtosecond laser. We further consider possible application of the sources designed as well as feasibility of scaling to the larger laser facilities.

This study was supported in its different parts by the RFBR (grants № 19-02-00104, 19-02-00740).

Simulation of the angular distribution of a laser-plasma X-ray source in picosecond region

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We performed a series of numerical PIC simulation for the theoretical interpretation of the spectral and angular characteristics of plasma X-ray radiation, measured on the Vulcan PW laser facility [1,2]. In particular, we carried out 2D numerical calculations of the interaction of intense laser pulse (intensity $\sim 5\times10^{20}$ W/cm^2, duration 0.7 ps) with a silicon foil 2–10 µm thick.

We investigated the effect of the laser pulse duration, as well as the effect of the prepulse and the target thickness on the bremsstrahlung X-ray angular distribution. Such optimization of the laser-plasma X-ray source may be important for the radiography or material science [3,4]. It was found that in the case of normal incidence of laser radiation on the target, the intensity of synchrotron radiation is about an order of magnitude higher, and the angular distribution of radiation is significantly anisotropic: most of the X-ray quanta are emitted along the normal from the front side of the target (Fig.1a.). For the case of 45°, no significant anisotropy of X-ray radiation was found (Fig. 1b). Note, that the efficiency of classical bremsstrahlung is approximately the same in both cases. It is known that electrons accelerated by the laser field are emitted along the target normal. Therefore, we assume that in the case of a normal incidence of a laser pulse, a spatial superposition of the reflected laser wave and a region of high concentration of electrons accelerated by the front of the laser pulse occurs, which leads to efficient generation of synchrotron X-ray radiation.

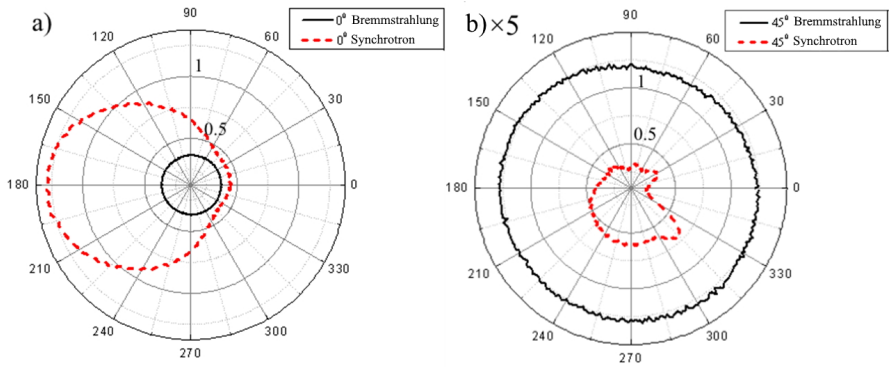

Figure 1: X-ray angular distribution. Silicon target, laser intensity was 3×10^{20} W/cm^2, pulse duration was 0.7 ps.

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Harmonics generation by atomic medium in optical vortex fields

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In the problem of the generation of high harmonics by an atomic gaseous medium there has now emerged an understanding that the description of the field incident on the medium in the scalar approximation does not allow solving the most complex problems of controlling the polarization of harmonics and to create more complex polarization state than elliptical, for instance, carrying optical vortex component. As a consequence, it is impossible to adequately describe the state of polarization of individual harmonics, as well as to localize the regions of generation of certain harmonics in a given state of polarization both in the direction along the optical axis and in the plane of the detector. Even in the case of interaction of the medium with a single-color sharply focused laser field in the focal plane the incident field is a complex object with field components in three Cartesian directions. The amplitude, phase and polarization properties of this field, if we go beyond the paraxial approximation of focusing, are fundamentally inhomogeneous both along the optical axis and in the focal and any other transverse plane. The theory formulated and developed in our group is a unique tool for describing such interaction, since is based on the solution of a truly three-dimensional problem of an atom in a field, the wave functions of which are specified not for a one-dimensional potential well, as in most theoretical works, but directly as wave functions of a hydrogen-like atom in spherical coordinates. Recently, many works, in particular those developed by our scientific group, have been devoted to the generation of high-order harmonics in an extended medium, taking into account the Gouy phase shift. The Gouy phase shift is the phase switching of the sharply focused radiation incident on the medium by an amount equal to π near the focal plane in a narrow layer. However, Gouy's phase shift is a theoretical phenomenon that arises when considering the focusing of light in the paraxial approximation of the wave equation. For a more rigorous description, it is necessary to use the vector focusing theories of Richards-Wolf [1] or Debye theory, which lead to vector field solution in the focal plane of a sharply focused beam.

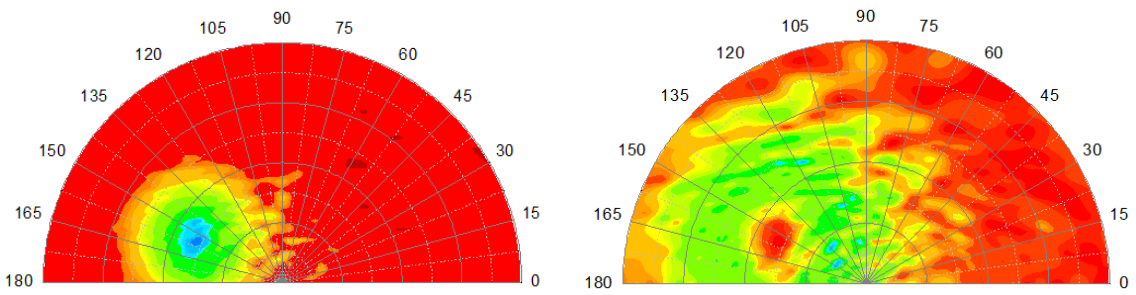

Figure 1: Transverse and longitudinal 3rd harmonic distribution at the distance 30λ from focal plane in the case of azimuthally polarized incident field.

In this work, on the basis of exact solutions for focusing Gaussian and non-Gaussian radiation (including radial, azimuthal and hybgid polarization states), with considering its longitudinal component, we investigate the question of the generation of harmonics in a thin rarefied medium localized near the focal plane, which make it possible to study the effect of the amplitude-phase features of the incident field on the process of generation of harmonics at the optical axis. In particular, the problem is posed of finding, for individual harmonics, the position of the longitudinal field maxima on the optical axis along the intensity and

determining the state of polarization in this region. We compare ellipticity diminution with focal plane distancing and focal spot form structuring for different modes.

At present, a number of experimental works have appeared, in particular [2-4], in which the question of the interaction of an atom with non-Gaussian field modes has been investigated. Non-Gaussian modes, in the case of achieving a high field intensity in them, represents a wide range of possibilities for controlling the polarization properties of harmonics, as well as obtaining "optical-free" focusing of harmonics, which is especially important for high harmonics. The most important result of the presented work is the appearance of asymmetry in the distribution of harmonics on the detector, which is especially pronounced in polarization-allowed measurements.

This work was supported by Russian Foundation for Basic Research (grant №18-02-40014).

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Indirect excitation of the ^{83}Kr low isomeric nuclear states using femtosecond laser plasma

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An excited state of a nucleus is called isomeric if it has a significant lifetime (¿ 1 ns) due to the strong suppression of the transition probability to lower states by the spin and parity exclusion rules. For example, nuclei ^{83}Kr have energy levels 9.4 and 41.6 keV with half-lives of 156.8 ns and 1.83 h, respectively. Isomeric levels find their application in precise calibration of xenon dark matter detectors (^{83m}Kr), medical radiography (^{99m}Tc), time and frequency standards (^{229m}Th). Due to the high energy-storage capabilities some materials with isomeric levels (^{178m2}Hf, ^{180m}Ta, ^{83m}Kr) are good candidates for the nuclear batteries or gamma-ray lasers projects. Nowadays, the issues of effective excitation and de-excitation of isomeric states are of interest.

Relativistic intensity femtosecond laser plasma is a brilliant source of ultra-short bursts of high energy electrons, ions, hard X- and γ-rays which can be used to excite isomeric nuclear levels. Usually, when using a laser plasma, *direct* isomeric levels excitation is considered due to mechanisms such as photoexcitation, inelastic electrons and ions scattering, nuclear excitation by electron capture (NEEC), nuclear excitation by electron transfer (NEET) etc. [1] This assumes the use of low-energy (10-100 keV) X-ray quanta, electrons or ions. In this work, we present a theoretical analysis of the efficiency for various excitation mechanisms using high-energy plasma radiation (¿ 1 MeV) [2]. It was shown that for the 9.4 and 41.6 keV levels of the ^{83}Kr nucleus, the *indirect* excitation scheme through short-lived levels with energies of 0.5–1.5 MeV is more efficient. Experimental schemes are proposed based on Kr cluster jet or high-pressure Kr chamber and solid target irradiated by relativistic femtosecond laser.

This work was supported by RFBR-Rosatom grant № 20-21-00030.

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Laser pulse focusing optimization in the case of nonlinear self-phase modulation

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Recently, great progress has been observed in nonlinear methods for the femtosecond laser systems peak power increasing based on post-compression nonlinear processing CafCA of laser pulses [1]. CafCA allows multiplying the peak power of the ultrashort laser pulse by bypassing the limitations associated with the diffraction gratings damage threshold. To date, the authors of this work have demonstrated a fivefold increase in the peak power of a laser pulse with an initial power of 250 TW [2].

At the same time, a multiple increase in power does not always correspond to an increase in peak intensity if focused. The nonlinear nature of CafCA leads to the formation of nonlinear phase distortions [1] caused by amplitude inhomogeneities in the near field of the radiation. Such distortion can lead to a significant decrease in focusing efficiency. Moreover, standard adaptive optical systems (for example [3]) are not capable of adequate treatment for nonlinear phase distortions. This is due to both the fundamental decrease in the focusability of the laser pulse and the difficulties in interpreting the data from the Shack-Hartmann wavefront sensor.

In this paper, we consider an approach that makes it possible to significantly improve the focusing of a pulse with nonlinear phase distortions that arose after CafCA using a bimorph mirror with a controllable shape of the surface. The approach is based on both a complex increase in the efficiency of adaptive wavefront correction in both linear and nonlinear cases.

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Phase space investigation of electron acceleration in DLA

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State-of-art laser systems are able to generate laser pulses of very short duration (10-1000 fs) with peak powers up to several PW. Due to various acceleration mechanisms, plasma electrons can gain the energy of the order of tens and even hundreds MeV. Direct laser electron acceleration (DLA) has been studied experimentally in the Laboratory of Relativistic Laser Plasma (Lomonosov Moscow State University, Russia) at 1 TW Ti:Sa laser system (peak laser intensity in vacuum 5×10^{18}W/cm^2). Collimated electron beams with characteristic divergence of 0.05 rad and temperature of 2 MeV had been observed [1].

Earlier results [1] of PIC simulation indicate that the electron acceleration in the plasma channel strongly depends on the initial parameters of the injected electron. In present work, we had carried out a detailed analysis of the electron acceleration process in the plasma channel to determine the optimal parameters of injected electrons, which is necessary to optimize the DLA regime in the experiment.

A simplified model of the DLA phase portraits was used [2], representing the electron energy versus the relative phase of the laser field and betatron oscillations, neglecting the rapidly oscillating part and the variability of the amplitude of oscillations of the electron transverse velocity. Changes in the topology and the nature of the bifurcations of phase portraits were analyzed depending on the amplitude of oscillations of the electron transverse velocity. A regime (topology of the phase portrait), in which it is possible to accelerate electrons with a low initial energy ($\gamma \sim 1$) to high energies $\gamma \sim 70$ ($a_0 = 1.5$), was found. The optimal values of the electron transverse velocity on the axis were obtained: $v_{xA} \sim (0.26 - 0.30)c$. Also, it was found that electrons with low initial energies are accelerated to a narrow energy range, which indicates the possibility of obtaining a mono-energetic electrons energy spectrum.

The same phase portraits were also obtained from the numerical integration of the equations of electron motion. The result qualitatively coincides with the earlier model, however it yields significantly higher maximum electron energies: $\gamma_{max} \sim 110 - 130$. The optimal length of the plasma channel ($\sim 150\lambda$) was also established.

The maximum energy γ_{max} gained by an electron in this acceleration regime was expectedly increasing with the increase of a_0, however, a deviation from the linear dependence obtained in [3] was observed.

This work was supported by RFBR grant № 19-32-60069. D. A. Gorlova acknowledges Foundation for the advancement of theoretical physics "BASIS" for the financial support.

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Perspective of ion acceleration with PW-ultrashort laser pulse

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The unique exploratory mission of this research is to build the scientific foundation needed to develop high energy laser-particle accelerators. After short survey of relevant background, this presentation will discuss the recent experimental findings on ion acceleration obtained on PW-ultrashort laser.

We will discuss the newly found scenario where ions are accelerated in the electrostatic field of charged cavity created by relativistic ultra-short and high contrast laser pulses at the target front and by the enhanced sheath field established by relativistic electrons at the target rear surface, via so-called enhanced TNSA mechanism. A "threshold" target thickness for proton acceleration is found. For targets thicker than the "threshold" the prepulse-induced preplasma at the target front can boost ion acceleration by increasing laser absorption while this is ineffective for thinner targets due to prepulse-induced plasma formation at the target rear. This dual nature of the preplasma is described analytically, and particle-in-cell simulations confirm this concept.

These investigations are closely related to recent development or imminently anticipated development of laser technology to bring the existing laser systems to a multi-PW level.

High-charge electron beams generation due to direct laser acceleration in subcritical plasma

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Modern high demand applications of relativistic laser-driven electrons such as formation of betatron source for phase contrast imaging, nuclear photonics require high repetition rate collimated high charge bunches of multi-MeV electrons. Up to date the most common electron sources operate in the laser wakefield acceleration scheme (LWFA) in low density ($10^{-4} - 10^{-2}$ of the critical value n_c) plasma providing relatively weak bunch charge of a few pC. Substantially higher charges (though with lower particles energies) may be generated in near-critical plasma in the regime of direct laser acceleration (DLA).

In this work it is demonstrated that injection of electrons can be reached with sufficiently dense but still subcritical (transparent for the laser radiation) micron scale thick plasma layer with subsequent DLA in the channel formed by the forward propagating pulse with $a_0 = 2$ at the rear side of the plasma slab. It is shown that such plasma configuration can be obtained by evaporation of $\sim$ 16 µm audio tape by an artificial nanosecond prepulse. The acceleration mechanism is combination of hybrid plasma instability and DLA. Numerical simulations demonstrating almost 50% injection efficiency are supported by experimental observation of electron bunch with divergency of $\sim$ 0.05 rad, charge of $>$ 1.6 MeV particles $\sim$ 50 pC using TW laser facility with the pulse energy as low as 30–50 mJ, hence reaching 1 nC/J efficiency.

Here we presented the simplest experimental realization of the proposed scheme. Electrons with energies up to 20 MeV could be obtained lengthening the channel to 30 µm as we saw in additional simulations. Such a configuration can be easily designed introducing a gas jet behind the thin film. Further increase in a_0 and/or pulse energy may provide for electrons with energies of 40–100 MeV and much higher bunch's charge. This path the way toward "table-top" compact high repetition rate laser accelerators for nuclear photonics and other applications.

Angular-spectral characteristics of synchrotron radiation generated in a regime of relativistic self-trapping laser propagation

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High-energy electron beams from laser-plasma accelerators can generate a bright synchrotron x-ray radiation due to betatron particle oscillations. A small size (down to 1 µm), ultrashort duration and relative low divergence make such betatron sources attractive for applications in biology, chemistry, medicine, and material science. At the same time in dense gas plasma a short laser pulse can propagate in relativistic self-trapping mode that provides effective conversion of laser energy to the accelerated electrons and maximizes the total charge of these particles, which emits a large amount of betatron radiation.

Based on 3D PIC simulations and test particle method, we have analyzed impact of a laser pulse on angular-spectral characteristics of x-rays in relativistic self-trapping regime. In this regime, radiation with 0.1-1 MeV photon energies, low divergence, and high brightness is generated. It has been shown that a 135 TW laser can produce up to 3×10^{10} photons of > 10 keV energy and a 1.2 PW laser makes it possible generating about 10^{12} photons in the same energy range. A laser pulse filling plasma cavity enables effective loading high number of electrons and triggers a soliton-like structure with strong accelerating plasma fields and rather long propagation distance. Interaction of particles with the laser pulse leads to generation of polarized x-rays, which have an anisotropic distribution in the plane perpendicular to the direction of the laser propagation. We predict extrabright x-ray beams, which can be produced with few J, tens of fs laser system working at 10Hz and might have order of magnitude larger (than already achieved) laser-to-photons conversion efficiency.

This work was supported by the Russian Science Foundation (grant no. 17-12-01283).

The analysis of the bremsstrahlung spectrum of the laser-plasma electron source in presence of pileup effect

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Recently, generation of up to petawatt short (5 to 50 fs) laser pulses, with intensities as high as 10^{18} W/cm^2 and more, became possible on a small-scale, tabletop systems. Such pulses cause acceleration of electrons upon interaction with plasma. This offers a possibility of creating tabletop laser accelerators which would have various advantages over conventional ones such as lower construction cost, lower maintenance cost, and compactness.

One of the techniques for the evaluation of the resulting accelerated electron pulse involves directing aforementioned electrons on the face of a dense target and measuring the resulting bremsstrahlung spectrum.

This method has an inherent ambiguity in the matter of spectrum transformation, as multiple processes take place simultaneously in the experimental setup. This can be mitigated, to an extent, by using numerical modelling by predicting the resulting gamma spectrum measurement with known electron spectrum and using this data in reverse, thus calculating the initial spectrum from the experimentally observed one.

Another problem that arises is the non-negligible probability of simultaneous registration of two gammas — the pile-up effect, which makes it difficult to process spectra with two (or more) parameters — such as, for example, the two-exponential spectrum, which is of great importance in the context of laser-plasma acceleration.

Both these problems are considered. An analytical analysis is performed, resulting in recommendations and techniques to be used in practice.

Section 2: Ultrafast phenomena in ionized gases

Section Chair:

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(Lebedev Physical Institute, Russia)

Program committee:

Alexander Popov
(Moscow State University, Russia)

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(Lebedev Physical Institute, Russia)

Scope

Instabilities and high-frequency phenomena in photoionized plasma

Non-linear phenomena in low temperature plasma
Kinetic processes in plasma

High intense unipolar THz pulses generation in nonequilibrium xenon plasma

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We propose the new approach to generate high intense unipolar pulses in terahertz frequency range. Based on self-consisted modelling of ultrashort CEP pulses propagation in xenon plasma while its fast relaxation we educed the specific conditions of proposed earlier gain regime [1,2] when the pulse undergoes nonuniform amplification within its length resulting in the rise of pulse leading edge while degradation of its trailing tail with the opposite polarity. The latter can lead to the formation of THz pulses with high degree of unipolarity. Simulations have shown that backward influence of propagated pulse on the plasma evolution brings to the saturation of gain regime. So, the gain drops while the pulse fluence is saturated at the level $\sim 10^{-5}$ J/cm^2. Thus, within the proposed amplification concept it is possible to obtain picosecond unipolar THz pulses with intensities up to several MW/cm^2. Both sine and cosine waveforms of one-cycle CEP pulse were considered to demonstrate the different scenario of unipolarity formation. The maximum obtained value of unipolarity factor $U = \frac{\int E(x)dx}{\int |E(x)|dx}$ is about 0.8 for the sine waveform pulse and up to 1 for the cosine one. We also note that the sign of unipolarity factor can change during the propagation and amplification process in dependence on the specific features of EVDF evolution resulting from the preferential amplification of different parts of the pulse. The generated unipolar pulses are characterized by wide plateau in the spectrum which makes them useful for the purposes of broadband terahertz spectroscopy.

This work was supported by the RF President Grant MK-1932.2020.2 and Foundation for the Advancement of Theoretical Physics and Mathematics ("Basis" Foundation) 20-1-3-40-1.

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Competition of low-frequency fields arising at the effect of the focused laser pulse on a conductor.

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The work is devoted to the study of low-frequency fields arising at the effect of an optical pulse focused into a strip on the conductor surface. The case of normal incidence of the laser pulse is considered. A general expression for the Fourier image of the low-frequency magnetic field in vacuum is analyzed in detail. It follows from this analysis that the total generated low-frequency field consists of two contributions. The first contribution, the quasi-cylindrical wave field, is the radiation field of a nonlinear source lying on the metal-dielectric interface. The second contribution is the surface wave field, which is the localized field propagating along the metal surface. The competition between these fields is studied. In the case when a few electron collisions take place during the characteristic time of the laser pulse effect, there is an area near the metal surface, the size of which is determined by the ratio of the real and imaginary parts of the permittivity at characteristic frequencies of low-frequency radiation, in which the surface wave is the main. In other areas near the metal surface, as well as far away from it, the quasi-cylindrical wave field is dominant.

Terahertz wave emission under action of a *p*-polarized laser pulse on plasma

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The generation of terahertz (THz) radiation is theoretically studied when the p-polarized laser pulse interacts with plasma [1]. The boundary problem for a p-polarized laser pulse falling on semi-bounded plasma is solved and it is established that at the plasma boundary the laser field increases significantly when laser pulse falls at the angle of total reflection. This effect of the laser field amplification is especially appears, when the pulse with short duration is incident at small angles on the near-critical plasma under conditions rare electron collisions. Based on the time averaged Maxwell equations and hydrodynamic equations, the excitation of THz fields in plasma caused by ponderomotive action of the laser pulse on electrons and their emission in a vacuum are studied. The spectral and energetic parameters of the THz signal are investigated as the function of the electron concentration, the laser pulse duration and incidence angle. THz wave packet energy density (fluence) was calculated. It is shown that the increase of the THz signal fluence occurs when laser radiation is incident at the angle of total reflection in two cases: a) for small incidence angles and b) for grazing incidence. Maximum of the THz pulse fluence at the grazing incidence occurs in a rarefied plasma, and similar to the case of the s-polarization [2]. It is shown that, at small incidence angles, the THz signal fluence is maximum for a near critical electron concentration and this maximum increases with decreasing duration of the laser pulse and frequency of electron collisions. In this case, the energy density of the THz signal for the p-polarized laser radiation is much higher than for s-polarization [2] if the laser pulse with short time duration falls at small angles on the near-critical plasma with rare electron collisions. The frequency characteristics of the THz radiation were studied under the conditions of maximum fluence at small incidence angles of the laser pulse and it was shown that the emission spectrum has the broad line at the frequency close to the double reciprocal duration of laser pulse. The time profile of the THz signal was studied and it was shown that the electromagnetic field in it has only one oscillation cycle during the time, which comparable to the laser pulse duration. The estimations show that under the action of p-polarized laser radiation for its almost normal incidence on the near critical plasma the generation of THz signals with high intensity and conversion rate can occur.

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High harmonic generation of an atomic system subjected in an intense IR field and XUV pulse: application to the spectroscopy and metrology of an attosecond pulses

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The big progress in the development of coherent light sources makes possible the generation of the laser pulses in a broad range of frequencies with ambitious characteristics in intensity, duration, and polarization. Depending on the carrier frequency, the intense laser pulse may in a different way interact with an atomic system: for example, the interaction of the XUV pulse with intensity $\sim 10^{16}$ W/cm^2 can be considered in the frame of perturbation theory, while mid-IR pulse with intensity $\sim 10^{14}$ W/cm^2 nonperturbative dynamics in the laser-mater interaction. Although both cases are well studied for monochromatic fields and generalized for the case of short pulses [1,2], the case of simultaneous interaction of both IR and XUV pulses has not been studied precisely.

In this work, we discuss the theoretical background for accurate analysis of the laser-mater interaction of short XUV and IR pulses. Our theoretical treatment is based on the developing perturbation theory in XUV pulse by using wave functions dressed (nonperturbative) by an intense IR field. We show that in the adiabatic limit, the IR-field-dressed wave function can be presented in the closed analytic form and be used further for the developing approximate expression for the Green function of an electron in an intense IR field.

We apply formal perturbative expansion in the XUV field for analysis of high harmonic generation (HHG) by an atom in an intense IR field assisted by a short XUV pulse. We show that perturbative XUV pulse induces new channels for HHG, which can be utilized for spectroscopy of two-photon transition matrix elements [3], tracing closed classical trajectories contributed to the HHG amplitude [4], and retrieving waveform of the XUV pulse [5]. We check the accuracy of our theoretical findings by making a comparison with results obtained from the numerical solution of the one-electron time-dependent Schrödinger equation as well as time-dependent Kohn-Sham equations modeling nonlinear dynamics of a multi-electron atomic system in an intense field.

The work is partly supported by RFBR (Grant No. 20-32-70213) and Russian Science Foundation (project No. 20-79-10322).

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Generation of THz radiation under the action of a focused femtosecond laser pulse on a semiconductor layer deposited on a metal surface

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Terahertz (THz) radiation generation in a semiconductor-metal nanostructure exposed to a femtosecond laser pulse is studied. Time dependence of the generated low-frequency magnetic field is found. In semiconductors transparent to THz radiation the magnetic field oscillations at the electron plasma frequency after switching off the laser pulse are observed. The spectral composition of the generated THz radiation pulse is found. The emission spectrum of transparent semiconductors has a peak at the electron plasma frequency. This peak is strongly broadened at high electron collision frequencies. The contribution from the plasma resonance region leads to a relative increase in the total energy of THz radiation. The effect of the laser pulse focusing on the THz radiation pattern was studied. The stronger focusing, the wider THz radiation pattern. It is shown that the optimal generation conditions are realized when the size of the focal spot is comparable to the skin depth and the pulse length. If the twice thickness of the semiconductor nanolayer is equal to an odd number of half-wavelenghts of high-frequency radiation the generation efficiency increases greatly due to the interference of incident and reflected waves in the dielectric. For GaAs and GaSb the generation efficiency increases by more than two orders of magnitude (see [1]).

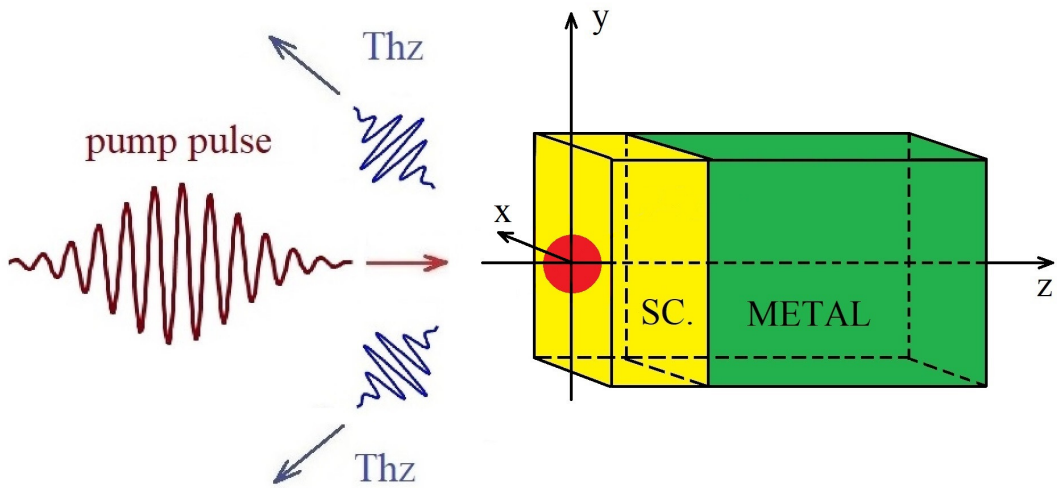

Figure 1: THz radiation generation scheme.

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Surface plasmon resonance enhanced second and third harmonic generation of intense near-IR femtosecond laser with periodic microtip metal structure

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Enhancing ultrafast nonlinear processes at a micro- and nanometer scale has the potential of creating sources of energetic particles with intense IR femtosecond lasers, useful for such applications as the development of directional IR photodetectors. Recently, on the basis of a polymer film with regular flogging, we have obtained a new type of metal near — surface 2D microstructures [1], which are micro-tips oriented normally to the substrate – a semi-infinite metal layer. This periodic metal nano- and microstructured targets prospective for IR frequency range in view of plasmon resonance condition production that leads to high energy coupling, enhancing local field accompanying increasing of optical harmonics generation efficiency.

The goal of this work is to theoretically and experimentally investigate dependence of absorbed energy and efficiency of harmonic generation using intense IR femtosecond laser radiation interacting in conditions of plasmonic resonance. In the framework of the developed theoretical model based considered the plasmon-polaritons surface waves [2] produced under interaction of focused femtosecond laser beam with the 2D grating on the metal wafer having constant period (2.9 µm) that supported with an accuracy of 10 nm. The height of tip varied from 0.5 µm to 10 µm. The height of the profile of a particular sample was assumed to be constant with an accuracy of 200 nm and was determined by the features of the synthesis of the structure. The surroundings permittivity is assumed as dielectric (air). Thank this model we find out that P polarized laser radiation near the plasmonic resonance spectral position leads to more efficient energy absorption and higher optical second and third harmonics yield.

The experiments used a femtosecond Cr:Forsterite laser (central wavelength 1240 nm, pulse duration 140 fs, energy 800 µJ). The radiation was focused on the sample by a lens with a focal length of 300 mm, the diameter of the focused beam was 120 microns, and the intensity on the target was about 5×10^{13} W/cm^2. The target was set at an angle close to the normal for the incident radiation. To collect the signal of the generated harmonics a spherical lens with a numerical aperture of NA = 0.78 and a focal length of 8 mm was used. Since the interaction with the target led to the generation of plasma on the surface and its partial damage, the target shifted when registering the spectra.

The experiments revealed the presence of signals of the second and third harmonics of femtosecond laser radiation (see Fig. 1). We also find that at intensity of 5×10^{13} W/cm^2 on the surface of the silver target with 3 µm tip height the second laser pulse lead to more efficient second harmonic generation signal, as predicted theoretically.

This work was supported by the RFBR grants № 18-29-20090, 18-02-40014.

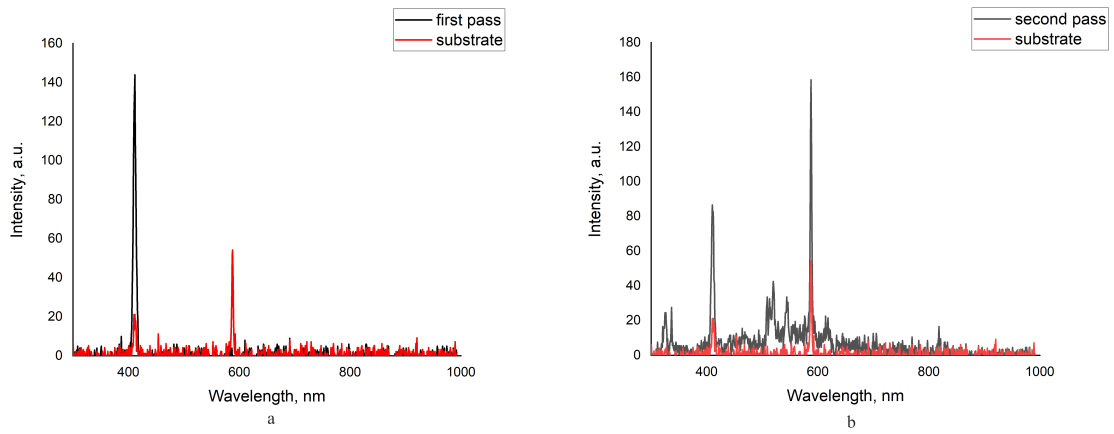

Figure 1: Spectra of the signal reflected from the target surface during interaction with the substrate (red curve), single (a) and repeated (b) impact on the structured surface (black curve).

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Analytical approach to ionization-induced multiwave mixing in multicolor fields

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The ionization of a medium by strong femtosecond two-color pulses is accompanied by generation of coherent secondary electromagnetic radiation. The generation is particularly manifested around various combination frequencies (CFs) of the two-color pump in different frequency ranges, from terahertz to the deep-ultraviolet, and can be interpreted as a result of the ionization-induced wave mixing. The main features of this phenomenon are governed by substantially high-order nature of nonlinear ionization and are manifested in very short (compared to pump) duration of generated pulses as well as in simultaneous generation of numerous CFs. In Refs. [1, 2], an analytical approach was presented and was used to calculate free-electron currents for linearly polarized two-color pump with one quasimonochromatic component being much weaker than the other. However, there is still a lack of systematic studies on the ionization-induced wavemixing in two-color fields in general case (with arbitrary polarizations of one-color components and arbitrary ratio between component intensities).

In this work, we present an analytical approach that should facilitate such studies. This approach allows one to calculate radiating currents generated by ionizing two-color femtosecond pulse at CFs in recollision-free regime. The method is based on the decomposition of ionization rate over quasimonochromatic components at various CFs and describes the excitation of electron current at any CF in a unified way (regardless of the particular frequency range). Using this ionization rate decomposition method, one can find the slow envelope of the current component at some CF as dependent on parameters of the ionizing two-color pump: intensities, polarizations, durations, and chirps of its one-color components as well as the phase and group shifts between one-color components. Knowing this dependences, one can simplify electrodynamical program codes or estimate analytically the conversion efficiency in some focusing models (for example, for short microplasmas from tightly focused pumps or for long quasi-uniform plasma columns from axicon-focused pumps).

The work was supported by the Russian Foundation for Basic Research (Grant No. 20-32-70213).

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Optical strong-field phenomena in crystalline solids driven by few-cycle mid-infrared laser pulses

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Nonresonant light-matter interaction can occur in several distinct regimes depending on the amplitude and frequency of the applied electromagnetic field. The border between the perturbative and strong-field regimes is determined by a dimensionless Keldysh parameter [1] relating the ponderomotive energy U_p of an electron-hole pair in the oscillating field to the width of the material band gap E_g. In the case of weak field ($U_p \ll E_g$), the interaction can be described using perturbation theory, which predicts multiphoton transitions and perturbative generation of harmonic frequencies due to high-order nonlinear optical susceptibilities. In contrast, when the driving field is strong ($U_p \geq E_g$), the mechanism of electron-hole production changes and quantum tunneling dominates. In this regime, nonperturbative phenomena such as generation of intraband currents, dynamical Bloch oscillations or dynamical Franz-Keldysh effect can be studied. Moreover, the properties of light-matter interaction depend on the polarization of the driving wave with respect to the crystallographic orientation of the studied material and when extremely short pulses with only few oscillations of the carrier wave are used, also on the carrier-envelope phase.

In this contribution I will review our recent experimental studies on the interaction of crystalline solids (diamond, silicon, 2D materials) with few-cycle mid-infrared laser pulses with stable carrier-envelope phase at the border between perturbative and nonperturbative interaction regimes. I will discuss three main types of experiments: i) polarization- and carrier-envelope phase-dependent reflective high harmonic generation in silicon and other materials, ii) impact ionization in diamond driven by mid-infrared pulses [2] and iii) valley-selective energy shift of exciton resonance in monolayer WSe_2 driven by strong nonresonant circularly-polarized fields.

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Mechanism of electron bunching by a relativistically intense laser pulse when crossing a nonuniform-plasma boundary

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The phenomenon of generation of attosecond electron bunches by a laser pulse of relativistic intensity interacting with a semi-bounded rarefied plasma of uniform density, which was discovered in [1], is numerically modeled in one-dimensional geometry. The aim of the study was to elucidate the features of the mechanism of electron beam generation, depending on the nature of the plasma boundary - whether it is sharp or blurred in the form of a transition layer separating the main volume of the plasma from the vacuum. It is shown that the nature of the plasma boundary significantly modifies the mechanism of beam generation, while preserving its main fundamental features [2-3]. It is found that in the case of a blurred boundary, the process of beam formation occurs in such a way that electrons from the boundary region of the plasma, in which the wake wave of the laser pulse is non-stationary, are captured into the generated beam. The electrons that are initially located in the transition layer are also involved in the formation of the generated beam. As a result, the generation of an electron beam is more intense, which contributes to a denser grouping of electrons in its head compared to a plasma with a sharp boundary. By cutting these electrons from the entire electron bundle by energy separation, a beam with characteristics superior to those obtained from a plasma with a sharp boundary can be obtained for a plasma with a diffuse boundary. By changing the plasma density and the transition layer length, as well as choosing the cut-off energy during electron separation, it is possible to control the parameters of the beam introduced into the laser-plasma accelerator [4].

The reported study was funded by RFBR and ROSATOM, project number 20-21-00150.

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Coherent control of the air molecule ionization and its Terahertz radiation with multiple color femtosecond pulses

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In this talk, I will present our recent work on the coherent control of ionization of air molecules by femtosecond laser pulses composed of 3 colors [1,2]. By variation of the phases $\Delta\varphi$ and $\Delta\gamma$ between the three frequencies, we observed significant change of the ionization degree of the oxygen and nitrogen molecules with modulation depth up to 20%. Furthermore, it is found that the dependence of the ionization on the two phases $\Delta\varphi$ and $\Delta\gamma$ changes with the laser intensity ratio between the 3 colors. Based on the concept of photonic mixing of the ionization, the observations are interpreted by the competition between different ionization channel.

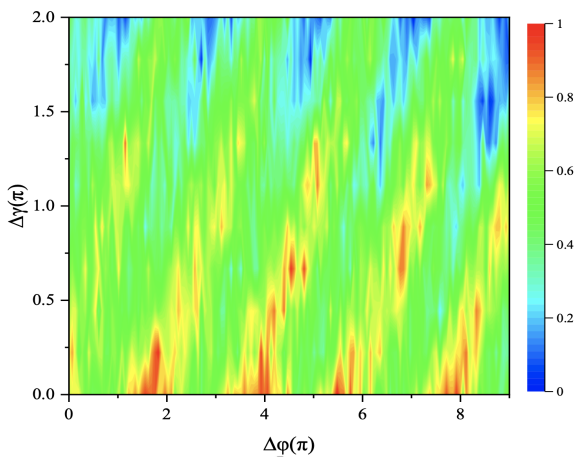

Figure 1: Fluorescence signal at 337 nm of the air plasma as a function of the phases $\Delta\varphi$ and $\Delta\gamma$.

In the meantime, the Terahertz radiation from the air plasma was studied simultaneously. It was found that the amplitude, polarity, and ellipticity of the THz radiation can be controlled coherently [1]. Simulations based on the photocurrent model reproduced well the observations. These findings provide a robust and simple method for enhancement and control of the ionization of air and its Terahertz radiation, which is of great significance for high-field physics.

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On the applicability limits of the sharp boundary assumption in the interaction of laser pulses with photoionised plasma

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The ionisation of gas atoms by a short pulse of laser radiation produces a plasma with a nonequilibrium distribution of photoelectrons. The density of electrons in such a plasma is inhomogeneously distributed in space. When electromagnetic field interacts with photoionised plasma the assumption of a sharp change in the electron density at the boundary is often used. In order to identify conditions under which this assumption is justified, we consider the interaction of monochromatic radiation with a plasma in which the electron density varies in a narrow layer.

We consider the situation when electromagnetic radiation with frequency ω falls normally on a weakly ionised semi-bounded plasma $z > 0$, produced by multiphoton ionisation of gas atoms. To describe the distribution of photoelectrons in plasma we used photoelectron distribution of the form $f(v, z) = n(z)/4\pi v_0^2 \delta(v - v_0)$, where v_0 - average speed of photoelectrons after ionisation, $n(z)$ - photoelectron density, that linearly increases from zero at the boundary $z = 0$ to the value $n(L) = n_0$ at $z = L$, then at $z > L$ remains constant. Solving the kinetic equation with the collision integral describing relaxation over the directions of photoelectron velocity without changing their energy and Maxwell equations, general expressions for electric field in plasma, reflection and absorption coefficients of incident radiation are obtained. Analysis of electromagnetic wave penetration features into plasma in high-frequency and normal skin-effects modes has been carried out. Comparing the obtained expressions with the results of [1], in which the plasma boundary was considered sharp, it is found that when the width of variable photoelectron density layer L is smaller than the skin layer depth corresponding to the penetration mode, the smearing of the plasma boundary does not affect the absorption and reflection coefficients.

The reported study was funded by RFBR according to the research project № 20-32-90158.

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Bolometric detection of broadband terahertz radiation by resonant heterostructures

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In this paper we analyze the possibility of constructing a bolometer capable of detecting terahertz pulses with an extremely large spectral width. Such a bolometric method of detecting was proposed in [1,2].

As noted in [1,2], penetration of electromagnetic signal into the conducting region of detector where the energy absorption occurs is crucial for bolometric method of detection. The penetration efficiency into this region can be increased significantly by placing dielectric layer under conductive layer. Such a dielectric layer can be considered as an electromagnetic resonator. If the frequency of the incident radiation coincides with the frequency of one of the resonator eigenmodes, a resonance of transparency is observed, which leads to effective filling of the resonator by the wave-field. This penetration of the radiation through the conducting layer is accomplished by the sharp increase of the energy absorbed in it. The proposed detector can be used to detect quasi-monochromatic radiation with the spectrum width does not exceeding the spectral width of the eigenmode. It is possible to increase the spectral width of the detector by replacing the conducting layer and the dielectric layer under it with a periodic structure consisting of a sequence of dielectric and conducting layers. Such heterostructure forms a photonic crystal with allowed and forbidden frequency gaps for electromagnetic radiation propagating within it. In this case, the operation range of the frequencies that we are able to detect using a bolometer will be determined by the width of allowed energy bands of the photonic crystal.

The possibility of broadband absorption of terahertz signals with the frequency range of about $\omega \sim 1$ THz with an efficiency of 50% based on the set of doped and undoped GaAs or Ge layers is discussed. Proposed detectors, in particular, can be applied for detecting of (sub-)picosecond pulses, as well as a sequence of pulses, the carrier frequency of which changes in time from pulse to pulse.

This work was supported by the Grant of the President of Russian Federation for State Support of Young Scientists (MK-1932.2020.2).

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Electromagnetic wave transformation in an abruptly created plasma layer

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We report on the transformation of p-polarized monochromatic plane wave under abrupt creation of a plasma layer and analyze the full mode structure of the transformed electromagnetic field. Such and similar configurations may occur when a semiconductor material is optically excited inside microwave or terahertz field [1–6]. Usually, they are studied as a basis for high-power microwave or terahertz switches, mirrors, and polarizers. The results of the present study show that the accompanying frequency conversion provides also broad possibilities for generation of strong frequency-tunable radiation at different frequencies. The tunability may be achieved through adjusting the angle of incidence and is facilitated by the rich spectral structure of the transformed radiation. The surface and leaky waves, transient radiation of continuous spectrum, and quasi-static fields are excited, and each of them may happen to contribute dominantly depending on the plasma parameters (plasma density, width of the plasma slab, and incident angle of the plane wave). The excitation of a leaky wave leads to formation of the prominent spectral peak around plasma frequency in the important situation corresponding to transformation of microwave radiation by photoconductive layer induced by a femtosecond pulse when the newly formed plasma layer is thin enough and plasma frequency is large compared to the frequency of the incident wave. Thus, the transformation may result in up-conversion from the microwave to terahertz range. Another peculiarity that the distinguishes the problem under consideration from similar problems is the excitation of the so called odd surface and leaky waves (with the transverse component of the electric field and the magnetic field changing sign in the middle of the plasma slab). This enriches the spectral and mode structure of the transformed field, providing possibilities for down-conversion and simultaneous generation of several spectral components.

This work was supported by the Russian Foundation for Basic Research (grant No. 20-32-70213).

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Effect of electron-ion collisions on the generation of terahertz radiation at the interaction of femtosecond pulse with plasma in magnetic field

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We discuss the possibility of a low-frequency radiation generation at a normal incidence of the laser pulse on a dense plasma in a constant magnetic field. The interaction of femtosecond laser pulse with dense semi-bounded plasma is considered under the conditions, where the plasma frequency of electrons is greater than the carrier frequency of radiation, i.e., laser radiation penetrates to the depth of a skin layer and the field strength exponentially decreases into the plasma. The ponderomotive force arising due to the field inhomogeneity is directed across the external magnetic field and varies during a time of the order of the laser pulse duration. Under the action of the magnetic field and a ponderomotive force, nonlinear low-frequency currents arise, including those along the plasma surface, which are the sources of low-frequency radiation. When exposed to a femtosecond pulse, the generated low-frequency radiation falls on the terahertz frequency range. Using the well-known ideas about the laser radiation penetration into a dense plasma and simplest equations for finding low-frequency currents arising at the ponderomotive action on magnetoactive plasma, we give a quantitative analysis of low-frequency radiation generation. Explicit dependences of generated low-frequency magnetic field versus the strength of external magnetic field, plasma parameters, and laser radiation are established, and the generated field strength is estimated [1]. The conditions are established under which collisions of electrons with ions affect the generation of terahertz radiation at normal incidence of a laser pulse on a dense plasma in a constant magnetic field. If, during the time of exposure to the laser pulse, not a small number of collisions of electrons occur, then the amplitude of the generated magnetic field decreases, and the total energy of terahertz radiation decreases. The shape of the generated pulse also changes, which can be used to obtain additional information on the frequencies of electron collisions. The influence of collisions on the generation of terahertz radiation manifests itself in a relatively weak magnetic field, when the electron cyclotron frequency is lower than the plasma frequency [2].

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3D modeling of short THz pulse propagation and amplification pulses in strongly nonequilibrium plasma channels formed in xenon by femtosecond UV laser pulses

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We consider the 3D numerical model based on the second-order wave equation for the propagation of a short THz pulse in a highly nonequilibrium plasma channel created in xenon by powerful KrF laser pulse of femtosecond duration. The effect of amplification of a THz pulse is achieved due to the formation of a photoionization peak in the spectrum of electrons located in the region of growing with the energy of the transport cross section [1]. We assumed that the THz pulse is rather weak and does not have an inverse effect on the electron energy distribution function (EEDF) in the channel plasma. We compare the 3D simulation and diffraction divergence of the pulse with the 1D model and analyze the dependence of the threshold of the pulse amplification on radius of the nonequlibrium plasma channel and initial size of the seed THz pulse. It is shown that the diffraction divergence results in the blue shift of the amplified THz pulse during its propagation. At gas pressures of several atmospheres, it is possible to achieve an increase in the energy of the THz signal by a several orders of magnitude at propagation length of ~ 30 cm.

This work was supported by Foundation for the Advancement of Theoretical Physics and Mathematics ("Basis" Foundation) 20-1-3-40-1.

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Generation of secondary radiation during interaction of intense laser field with fullerenes

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Strong-field phenomena during the interaction of intense laser pulses with gases and solids attract significant interest from the scientific and applied points of view. The increased interest in these phenomena is associated with the possibility of realizing effective methods for generating coherent broadband electromagnetic radiation in hard-to-reach frequency ranges. These phenomena include the generation of high-order harmonics of the ionizing laser field [1, 2], as well as the excitation of a low-frequency current in the plasma generated by a laser pulse, which can lead, in particular, to the generation of coherent terahertz and mid-IR radiation [3-6].

A gas consisting of fullerenes is an attractive medium for the generation of secondary radiation [7, 8]. In particular, this is due to a large number of electrons actively interacting with an ionizing pulse and related capability to obtain high values of emitting electron currents. In this paper, we study the interaction of C_{60} fullerene with a two-color laser pulse with a frequency ratio close to two. For this case, the efficient generation of secondary radiation occurs at a detuning frequency of a higher frequency from the doubled lower frequency of the two-color pulse [4-6]. To calculate the excited electron current, we numerically solve the three-dimensional time-dependent Kohn-Sham equations. Based on the comparison of the results obtained for fullerene with the results for the hydrogen atom based on the time-dependent Schrödinger equation, the differences in the mechanisms of the low-frequency current generation are analyzed. It is shown that the generation of mid-IR radiation at the detuning frequency in C_{60} fullerenes is more than an order of magnitude higher than for hydrogen atoms at intensities, corresponding to the same degree of ionization.

The work is partly supported by RFBR (Grant No. 20-32-70213).

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Retrieving of an attosecond pulse waveform based on the high harmonic generation

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We propose a new method for retrieving an attosecond pulse waveform based on the analysis of high harmonic generation (HHG) in an intense infrared (IR) field and a weak (perturbative) time-delayed extreme ultraviolet (XUV) attosecond pulse. The theoretical background of the proposed method is based on the recently developed approach [1], which treats electron-laser interaction with an intense IR field quasiclassically (i.e., in terms of closed classical trajectories), while interaction with attosecond XUV pulse is considered within perturbation theory. Generally, the XUV field may induce several alternative channels for HHG, however only one contributes for harmonics beyond the cutoff of IR-induced plateau. In this channel, electron tunnels through the barrier and accelerated by an intense IR field along a closed classical trajectory. At the returning moment $t = t_j$, the liberated electron recombines to the initial state with simultaneous absorbtion XUV photon with frequency Ω and emission harmonic of frequency Ω_h forming additional plateau in the HHG spectrum [2]. Our theoretical analysis shows that contribution of this channel causes a specific dependence of HHG yield on the time delay between IR and XUV pulses, which can be utilized for retrieving attosecond pulse wave form:

$$\mathcal{Y}(\Omega_h) \approx \mathcal{Y}_0(\Omega_h) + F_{\mathrm{XUV}} \sum_j a_j f(t_j - \tau) \cos(\Omega(t_j - \tau) + \phi_j) + F_{XUV}^2 \sum_j y_j f^2(t_j - \tau), \quad (1)$$

where $\mathcal{Y}_0(\Omega_h)$ is the HHG yield in an intense IR field, F_{XUV} and $f(t)$ are field strength and envelope of the attopulse, τ is the time delay between IR and XUV pulses, a_j, y_j and ϕ_j are functions of IR field parameters. The first and second terms contribute on the slope of the IR-induced plateau, while the last term in Eq. (1) dominates on the XUV-induced plateau [2]. According to the explicit expression for the HHG yield we suggest two procedures for the attopulse waveform retrieving: (i) using harmonics just below the cutoff of IR-induced plateau; (ii) using harmonic far beyond IR-induced plateau. These methods consist in measurement of the HHG yield as a function of the time delay. In the first case, the τ-dependence of $\mathcal{Y}(\Omega_h)$ provides full temporal characterization of the attosecond pulse waveform, while in the second case measurements of HHG yield mimic the square of attosecond pulse envelope. Practical realization of the proposed method requires that the difference between two recombination times should be larger than duration of the attosecond pulse. In order to test our method we perform numerical simulation of HHG in the presence of attosecond pulse within our recently developed time-dependent Khon-Shem code [3]. Our numerical calculations for Ne atom demonstrate the stability and high accuracy of suggested retrieval method of the waveform of the attosecond pulse.

The work is partly supported by RFBR (Grant No. 20-32-70213).

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Multielectron effects in high-harmonic generation by atoms in intense laser field

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The high-harmonic generation (HHG) of intense laser pulses is one of the most well-known phenomena of strong-field physics, which is of great interest in terms of numerous applications [1,2]. The physical mechanism of HHG is qualitatively explained in the framework of a three-step scenario [3]. At the first step, atoms or molecules are ionized by an intense field. At the second step, the electric field accelerates the free electrons in the continuum. At the final step, the electrons recombine with the emission of high-energy photons in a wide energy range. In this paper, we present our recent results on studying the multielectron effects at the first and final steps of HHG in atomic gases. To study the influence of multielectron dynamics on HHG, we have developed the computer code for the numerical solution of the 3D time-dependent Kohn-Sham equations for atomic orbitals, which takes into account the interaction of electrons with the laser pulse, nucleus, and each other [4].

Our numerical simulation for argon, krypton, and xenon has demonstrated that a significant multielectron effect in HHG at high laser-pulse intensity is the laser-induced polarization of the atom itself. The structure of the polarization potential at the atomic scale corresponds to decreasing the external electric field, decreasing the ionization probability, and suppressing the depletion of atomic orbitals. The latter can lead to a significant increase in the intensity at which the HHG yield reaches saturation and, correspondingly, to a substantial increase in the HHG yield at high laser-pulse intensities [4].

The second direction of research is devoted to the multielectron effects that take place when electrons recombine with parent ions [5]. We have studied the HHG in xenon and shown that perturbation of the electron-electron interaction potential induced by recolliding photoelectron wavepacket originated from the $5p_0$ orbital leads to the collective oscillations of all orbitals on the 4th shell closely localized in space and strongly interacting with each other. The resulting HHG yield is enhanced by more than an order of magnitude compared with the response of the single $5p_0$ orbital in the energy range near 100 eV. The high accuracy of the numerical results is confirmed by comparing the calculated HHG spectra with experimental results [6] and an analytical parameterization of the HHG yield.

The work is partly supported by RFBR (Grant No. 20-32-70213).

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Potential instability of collision plasma formed by multiphoton ionization of inert gas

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During multiphoton ionization of gas atoms by short pulse of linearly polarized laser radiation, weakly ionized plasma is formed. After switching off the pulse, at times of the order of the reciprocal frequency of electrons collisions with neutral atoms ν^{-1}, the non-equilibrium isotropic velocity distribution of photoelectrons is formed. This distribution has the form of a narrow peak in the energy region of the order of 1 eV. For such a plasma, along with high-frequency plasma wave, the possibility of the existence of a new unstable longitudinal electron mode in the long-wavelength region is predicted. The growth rate γ of this mode does not exceed half of the electron Langmuir frequency ω_L. The appearance of such a mode is associated with the features of photoelectrons elastic scattering by neutral gas atoms, arise as a result of Ramsauer-Townsend effect which is typical for inert gases. This effect consists in an anomalous decrease in the transport cross section of photoelectrons elastic scattering by atoms in the energy range characteristic for multiphoton ionization.

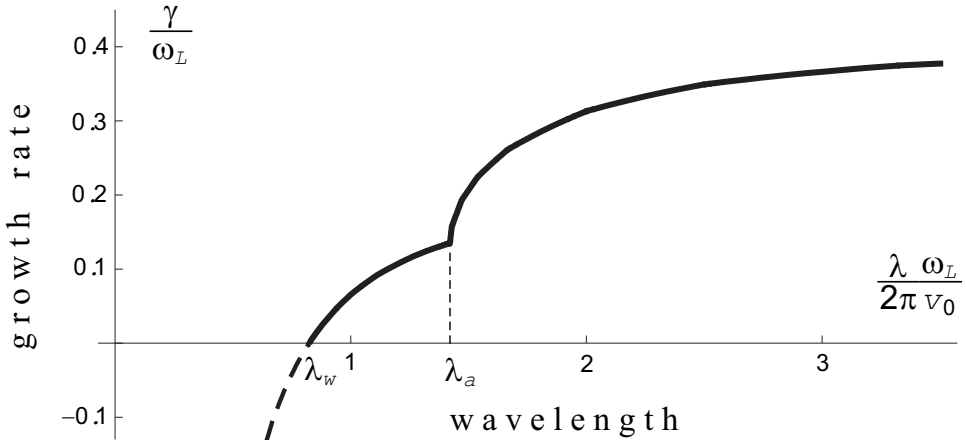

Figure 1: The dependence of instability growth rate on mode wavelength in strong collision photoionized Xe plasma with the ratio $\nu/\omega_L = 1.5$. Here v_0 is the photoelectron velocity.

Figure 1 shows instability growth rate of new mode as a function of wavelength of electron perturbations in plasma of Xe. In long wavelength region $\lambda > \lambda_a$, the amplitude of the perturbations increases aperiodically in time. In inter-mediate wavelength interval $\lambda_w < \lambda < \lambda_a$, the obtained mode is transformed into a longitudinal unstable wave. In short wavelength region $\lambda < \lambda_w$, the described mode is strongly damped. Note that when the collision frequency of photoelectrons is not anomalously low, the ordinary plasma waves are strongly damped in the most interesting long wavelength region, and the plasma becomes unstable with respect to low-frequency potential instability development.

For typical parameters of the considered object, the growth rate of the described instability lies within the THz frequency range.

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Section 3: Ultrafast phenomena in condensed matter

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Scope

Ultrafast optical, electronic and hot-carrier dynamics in metals and semiconductors

Ultrafast electron-phonon relaxation and phase transitions

Femtosecond-laser ablation and nanofabrication

Dynamics of ultrafast melting in laser-irradiated ruthenium thin films

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Ruthenium (Ru) thin films are used as grazing incidence mirrors in XUV and X-ray optical systems. Under sufficiently high fluences, irradiation by a pulsed laser can cause melting of Ru films. The melting and recrystallization may lead to defect formation and degradation of a film. The process of ultrafast melting, or even the melting threshold of Ru, are not yet extensively explored and understood.

In the case of femtosecond pulses under grazing incidence, the absorbed energy distributions are similar for a wide range of photon energies, which results in a similar thermal response of a system to irradiation [1]. Exploiting this fact, for the study of ultrafast melting one can apply more accessible visible light lasers.

In this work, we present an experimental and theoretical study of single-shot melting threshold in Ru films with thicknesses up to 100 nm. We applied the pump-probe technique to measure the transient change of reflectivity of Ru thin films on Si substrate after irradiation with 800 nm femtosecond laser pulses with varying incident fluence. The observed saturation of the reflectivity change as a function of the laser fluence was considered as an indication of the onset of melting.

Theoretical analysis of the changes of optical and electronic properties in the irradiated Ru target was performed using the hybrid simulation tool XTANT-3 [2]. It revealed that after reaching melting, optical properties of Ru are not sensitive to further increase of the laser fluence, which confirms the performed experimental approach to the melting threshold determination. For additional analysis, we also applied the two-temperature model, which traces changes in electron and lattice temperatures. The obtained threshold fluences in both models coincide, which serves as an additional validation of the performed analysis.

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Femtosecond optical breakdown of silicon

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Photoionization, energy deposition and plasma formation in crystalline silicon irradiated by intense near infrared laser pulses with varying pulse duration < 100 fs were theoretically investigated. Ultrafast optical breakdown with threshold energy fluence $\Phi_{th} > 1$ J/cm^2 was established by the sudden increase of the absorbed laser energy inside the bulk. The optical breakdown is accompanied by severe spectral broadening of the transmitted pulse. For the studied irradiation conditions, we find that the threshold fluence increases linearly with the increase of the pulse duration, while the corresponding laser intensity threshold decreases. The effect of the high plasma density on the stability of diamond lattice is also examined.

Optical frequency combs from quantum cascade lasers

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In this talk I will discuss how resonant light-matter interaction in the gain medium of quantum cascade lasers gives rise to a rich nonlinear multimode dynamics and a variety of phase-locked multimode regimes, most notably optical frequency combs with separation between the comb lines changing from one to many dozen round-trip frequencies. I will review recent progress in understanding why frequency combs seem to be so ubiquitous in QCLs and their emerging applications.

A standard method of producing frequency combs is based on mode-locked lasers generating ultrashort pulses, which operate mostly in the near-infrared and visible ranges. The mid-infrared (mid-IR) and terahertz (THz) spectral regions, where most chemical compounds have strong spectral fingerprints, hold enormous potential for frequency comb applications. Quantum cascade laser (QCLs) cover most of the mid-IR and parts of the THz spectral regions. Unfortunately, ultrafast gain relaxation in QCLs effectively prohibits ultrashort pulse generation through passive mode locking. Active mode locking schemes are still feasible but they are less convenient and produce pulses of limited peak power.

That is why the realization that strong resonant nonlinearity of the gain transition itself is enough to trigger the self-starting formation of frequency combs in QCLs was such a pleasant surprise. However, unlike the shorter-wavelength combs generated by ultrashort-pulse lasers, in QCLs the underlying periodic modulation is linked to a predominantly frequency-modulated optical wave. Still, the amplitude modulation due to population pulsations is always present to some extent and can be directly measured. This is especially true for the harmonic combs in which the lasing modes are separated in frequency domain by a large number of free spectral ranges. The harmonic combs were found to be self-starting too in all kinds of QCLs. The separation between neighboring lasing modes in the harmonic combs reaches hundreds of gigahertz and even THz frequencies, which implies coherent (sub)picosecond intracavity modulation. Therefore, harmonic QCL combs provide direct link between mid-IR lasing and coherent (sub)THz generation and modulation, which makes them promising as (sub)THz coherent sources and transceivers. Many aspects of the underlying physical mechanisms which give rise to complete suppression of neighboring cavity modes and the harmonic comb formation remain a mystery. Yet another recent puzzle is the formation of frequency combs in ring QCLs that were supposed to be single-mode. It is fascinating how QCLs continue bringing new surprising features to such a well-studied field as fundamental laser dynamics.

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Dynamics of propagating and localized excitations across interfaces analyzed by femtosecond solid state spectroscopy

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An excited electron propagates in condensed matter with its momentum $\mathbf{k}$ at an energy $E(\mathbf{k})$ and experiences elastic and inelastic scattering processes, which lead to electronic relaxation and energy transfer to microscopic excitations of the lattice and spin systems. Experiments employing femtosecond time-resolved photoelectron spectroscopy exploited so far very successfully the surface sensitivity of the method and probed such scattering processes locally at the surface or the surface near region in the time domain [1]. Here, we report on experimental results which analyze the non-local dynamics of excited electrons in two-photon photoemission (2PPE) and demonstrate sensitivity to buried media [2]. In these experiments one photon excites in Au/Fe/MgO(001) heterostructures electrons in Fe. Electron propagation through the layer stack to the Au surface is detected in 2PPE in back side pump front-probe experiments in a time-of-flight like scheme. We observe pronounced differences between front and back side pumping of the heterostructure which are attributed to electron transport contributions through the layer stack. Furthermore, competition of e-e with e-ph scattering will be discussed in $[\mathrm{Fe/MgO}]_n$ heterostructures. Pump-probe experiments of element specific spectroscopy in combination with electron diffraction provide here unprecedented insights regarding the mechanism of energy transfer across interfaces and emphasize the importance of coupling of electrons to non-thermalized interface phonons [3]. Extension of these experimental tools to address effects of strong electron correlation [4] and spin-dependent dynamics across interfaces [5] will be discussed.

This work was funded by the Deutsche Forschungsgemeinschaft through the Collaborative Research Center CRC 1242.

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Dual wavelength ultrashort laser ablation and surface modification of silicon

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

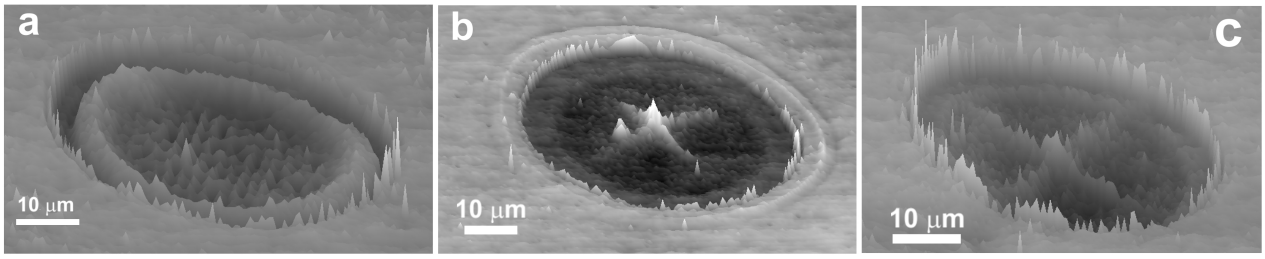

Figure 1: Confocal 3D images of spots produced by visible and IR pulses of equal energies with delay of -30 ps (a) and 0 ps (b) and by only single 515-nm pulse (c). The total fluence is 1.6 J/cm^2.

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Effects of laser-induced stresses in material modification

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More than 60 years passed since the first laser, one of the most impressive inventions of 20th century, shined its ray to the world. During this time, lasers have become an instrument without which it is not possible to imagine both everyday life and many fields of scientific research and industrial applications. However, a great variety of existing and emerging lasers with different irradiation parameters and a variety of materials, which can be treated by lasers for improving/modifying their properties or for industrial needs (cutting, drilling, cleaning, etc.), imply a wealth of processes contributing to the laser-matter interaction phenomenon that makes it to be still a hot topic of research.

Laser-solid interaction starts from light absorption by material after which the irradiated matter evolves thermodynamically through a sequence of non-equilibrium and quasi-equilibrium states to its final (laser processed) structure. Heating, melting, ablation, resolidification can occur differently even of the same sample, depending on laser irradiation parameters (wavelength, pulse duration, energy density, repetition rate). At short and ultrashort laser pulses, when laser-induced heating of materials in a surface layer or in the bulk is highly localized, strong temperature and, hence, pressure gradients are created, which affect post-irradiation evolution of matter via generation of stressed states and stress waves [1,2]. In the cases of thin films and layered structures (laser-processed solar elements, laser-induced forward transfer techniques, etc.), stresses may play a determining role in material ejection from the laser-irradiated samples [3-5].

In this talk we will discuss the role of stresses in material evolution during and after laser irradiation at femto-, pico- and nanosecond pulse durations and different wavelengths from UV to mid-IR. Examples of numerical simulations for metals, semiconductors, and dielectrics, based on specific models, will be demonstrated in both surface ablation and volumetric confinement regimes. The role of laser-induced stresses in triggering the explosive solid state crystallization of amorphous semiconductors will be discussed. It will be shown that, in many cases, for producing the desired material modification/ablation/deposition, control over the laser-induced stresses is necessary that can be achieved via tailoring the laser energy coupling.

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Broadband antireflection of GaSe crystals for optical para- metric oscillators

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GaSe crystals demonstrate the excellent combination of parameters for efficient optical frequency conversion and detection over a large range of wavelengths. However, a high refractive index (average $n = 2.63$ in the 2–16 µm range) leads to significant reflection losses (Fresnel losses), thus limiting conversion effectiveness.

The conventional method to overcome the Fresnel losses issue relies on the usage of multi-layer antireflection coatings (ARCs). However, mid-IR broadband multi-layer ARCs cannot be reliably applied to GaSe surfaces [1]. This is due to difficulties in growing single-crystal samples. It is challenging to achieve surface quality sufficient to be reliably coated with thick mid-IR multilayers coating. Even though it is possible to produce optical quality surfaces along the (0001) direction, access to other crystallographic directions and fine polishing of samples are hampered because GaSe has very low hardness. Moreover, the optical quality of grown GaSe crystals is limited by point defects and micro-defects. As a result, the physical properties of GaSe depend on the growing technology and the experimental conditions, which differ from one supplier of GaSe to another [1].

As an alternative, the fabrication of antireflection microstructures (ARMs) on the crystal surface can be used to reduce Fresnel losses. ARM represents a system of micro-sized features spaced on the sample surface. The antireflection effect is provided by smoothing the transition boundary between media and substrate and is can be described by the effective medium theory [2,3]. In past decades numerous methods have been developed for ARM fabrication [3,4]. However, only a limited quantity of them are applicable in this case, because the majority of ARM fabrication technologies rely on the surface quality of the substrate, e.g. to deposit a pre-fabrication mask [4].

We have already demonstrated the possibility of applying ARM to a GaSe surface using surface quality independent direct femtosecond laser ablation method [5]. Here we present new results of our experimental research towards broadband antireflection for GaSe nonlinear frequency converters.

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Nanoscale topographical, structural and chemical patterning of surfaces by ultrafast laser irradiation

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Ultrafast light coupling to metallic surface and band-gap materials enables manufacturing at the nanoscale that may trigger novel optical, fluidic and biomedical applications. Driven by remarkable surface material response to laser photoexcitation implying extreme states of temperature and pressure far from equilibrium, organized and self-organized patterns can be formed on the irradiated surface [1-2]. In addition to topography, design of periodic structures with arranged structural state offers new ways to manipulate material properties at the nanoscale and can be realized by spontaneous ordering of matter induced by laser irradiation [3]. In that respect, phase transition localized by the ultrafast photo-excitation and controlled by the coherent light polarization enables to alternate crystal-amorphous structures on the same area. Material periodically experiencing extreme non-equilibrium conditions opens new perspectives to create unconventional combination of allotropic phases to upgrade and even revolutionize existing surface functionalities.

From a fundamental perspective, complex surface topography is driven by the regulation that originates from the constructive and destructive roles of the electromagnetic and hydrodynamic processes. During the lifetime of a transient laser-induced melting layer, we uncover that dissipative structures can emerge and continuously exchange energy and entropy with photon flux to ensure relief growth and topographic organization upon solidification. Self-organized nanopatterns with various symmetries and aspect-ratios can be formed, demonstrating the potential of temporal beam shaping for the fabrication of advanced surfaces with a topography designed at the nanoscale. Upon multi-pulse excitation, the roles of collective effects and inter-pulse feedback on the resulting surface topographies will be presented by combining microscopy measurements to 3D numerical approach [4]. This couples electromagnetism to hydrodynamics in order to simulate the hydrodynamic flows and instabilities, that drives the matter towards self-organized convection nanocells, potentially induce thermocapillary waves oriented by nonradiative sub-diffraction near-field patterns.

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

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Last decade, understanding of transient excitation of electrons in solids has brought important developments for several classes of materials, at the level of both fundamentals and applications. While laser-excitation of dielectrics induces measurable ultrafast currents in the PHz regime [1], employing few-cycle laser pulses with controlled carrier envelope phase enables a coherent control of the electron dynamics with reduced crystal damage probability [2,3]. Notably, the possibility of transiently closing the band-gap of solids during their irradiation by linearly polarized ultrashort laser pulses was evidenced and attributed to the light-induced Zener tunneling [3-5].

The corresponding ultrafast modification of the band structure induced by laser dressing of electronic states can be measured experimentally [6,7] and analyzed theoretically using the Floquet formalism [8,9]. Despite its simplicity and limitations, it is applicable to several classes of materials [6,7,10] and enables to study the effects of light coupling with electrons in solids for a wide range of experimental conditions at a reduced computational cost [11].

In this work, after preparing the electronic band structures of two metals (Au and Mo), a semiconductor (Si) and a dielectric (α-SiO_2) using the density functional theory (DFT), the effects of dressing by a polarized laser light on the corresponding electronic band structures were investigated using the Floquet formalism [8,9,11]. While a selective excitation of the electrons can be achieved via a choice of laser wavelength and field strength [9,11], the Floquet simulations illustrate how the change in crystal orientation [12-15] and laser polarization [16] affects the electron dynamics in several solids.

Overall, the proposed approach outlines promising ways for selecting materials and laser parameters, via a cost-effective computer-aided manner, broadening perspectives in ultrafast photonics [6,17].

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Laser generated nanoparticle decoration of polymer micro powders for laser powder bed fusion

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Additive manufacturing techniques like laser powder bed fusion (LPBF) allow key advantages in the fabrication of objects and parts as custom design and fast processing. However, the library of available polymer materials for LPBF is still limited due to processing requirements as an optimum flowability or sufficient absorption of the laser energy. Besides, the functionalities of the generated parts are limited to the properties of the employed polymer powders. To address those problems, the addition of laser generated nanoparticles to the polymer micro particles is proposed, Fig 1a.

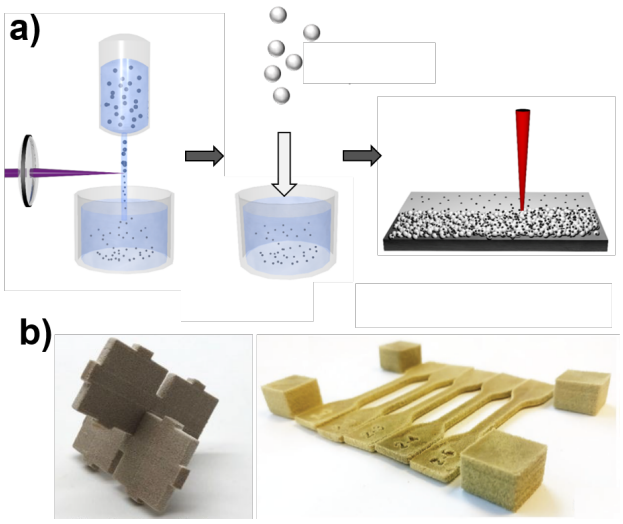

Figure 1: a) Schematic procedure employed for the generation, supporting and LPBF processing of nanoadditivated polymer powders. b) LPBF generated parts employing the nanoadditivated polymer powders.

The process is proved successful for different nanoparticle types (silver, carbon, iron oxide...), achieving the controlled modification of the optical and/or magnetic properties of the nano-decorated powders and transferring those properties to the generated parts by LPBF, Fig. 1b [1,2].

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Thermochemical LIPSS formation on Ti, Hf films at various ambient atmospheres

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Laser-induced periodic surface structures (LIPSS) have been the subject of intensive research in the last twenty years [1]. Surface relief of LIPSS is formed under impact of the focused laser radiation and observed on metals, semiconductors, and dielectrics. Relatively recently, thermochemical LIPSS (TLIPSS) have become the subject of active research due to a high regularity of the structures with period approximately equals to or less than the wavelength of laser radiation [2]. TLIPSS are formed due to the oxidation of the material surface illuminated with ultrashort laser pulses and are characterized by the elevation of the relief, forming parallel oxide protrusions. Formation of these structures by fs laser pulses on the surface of titanium films in the air was studied previously [3]. However, the lack of oxygen and high content of nitrogen will significantly influence the formation process which needs to be investigated. Here we present the results of the TLIPSS fabrication on a Si-Ti bilayer and Si-Hf films sputtered on the glass substrate using an astigmatic Gaussian IR femtosecond beam with a diameter of ≈150 µm along the major axis both in air and a nitrogen-rich atmosphere. Periodic structures with various morphology depending on scanning speed, laser power, and pulse repetition rate were obtained. Low-ordered ablative LIPSS with orientation perpendicular to the polarization are formed at high power levels and low pulse repetition rate in a nitrogen-rich atmosphere. The formation of ordered TLIPSS with period of ≈950 nm oriented parallel to the polarization is observed for slow scanning speeds (∼1 µm/s) and low pulse energies in a nitrogen-rich atmosphere. Raman spectroscopy analysis of these structures showed the presence of TiO_2 peaks (440 and 607 cm^{-1}), as well as Raman signal at 230 cm^{-1} and 552 cm^{-1} that are close to the frequencies of TiN (242 and 262 cm^{-1}).

This work was supported by the Russian Science Foundation (Grant No. 21-72-20162).

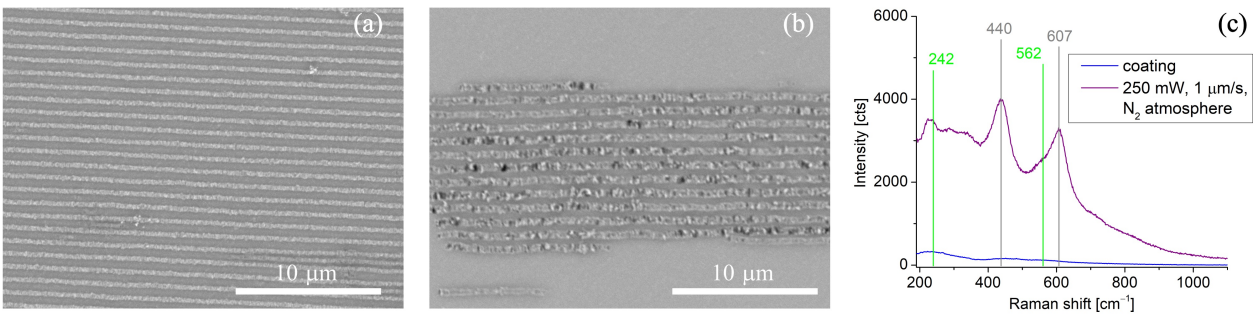

Figure 1: The SEM images of the TLIPSS formed on Ti film in air (a) and N_2 atmosphere (b). Raman spectra of the TLIPSS formed in N_2 atmosphere.

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Ultrafast excitation and detection of laser-driven two-magnon dynamics in a Heisenberg antiferromagnet with anisotropy

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The use of femtosecond laser pulses offers new possibilities to control spins in materials through affecting magnetic-dipolar, spin-orbital [1], and even exchange interactions governing magnetic properties [2-4]. Interestingly, even a weak perturbation of the exchange interaction can trigger spin dynamics which involves magnon modes at the edge of the Brillouin zone, possessing ultimately short wavelengths and energies in the range of 10-100 THz [5-7]. However, existing experimental observations [1,2] and theoretical descriptions [3,6] of this ultrafast regime of spin dynamics leave a number of open questions, impeding interpretation of experimental results and limiting further development of exchange-driven coherent spin dynamics.

Here we report on a theoretical analysis of laser-induced two-magnon excitation in a dielectric Heisenberg antiferromagnet with cubic structure and weak magnetic anisotropy. Excitation with a few fs short laser pulse was described as an impulsive perturbation of the exchange integral depending on the laser pulse polarization. We used Holstein-Primakoff and Bogoliubov transformations to diagonalize the Heisenberg Hamiltonian and calculated temporal evolution spin correlations in response to such perturbation. As the most representative cases we considered Néel vector and easy-axis aligned either along cube diagonal or along bonds between nearest neighbors.

We observed that the considered perturbation results in excitation of a two-magnon mode seen as an oscillating spin correlation function. The highest amplitude of the laser-driven two-magnon mode can be obtained with the electric field of the laser pulse directed along the bond between nearest neighbor spins. Importantly, we found that the time-dependent spin correlations perturb the Néel vector, i.e. contribute to macroscopic magnetic dynamics, only if the medium possesses magnetic anisotropy.

To reveal how such nontrivial dynamics of spin correlations can be detected in experiment, we analysed the corresponding changes of the dielectric permittivity tensor. We show that time-dependent spin correlations can have a two-fold impact on optical properties, either through direct modulation of tensor components or indirectly through modulation of magnetic birefringence proportional to the length of the Néel vector. The results of theoretical analysis are compared with the available experimental data [6-7].

This work was partly supported by the Foundation “BASIS” (grant № 20-1-5-95-1) and EU COST Action CA17123 “Magnetofon”.

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Ultrafast laser micro- and nano-modification of metallic bilayer thin films

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Results concerning the laser modification of bimetallic thin films (BTFs) by ultrafast (ULF) laser irradiations are presented. Experimental samples, made up of alternate titanium and aluminum or nickel layers, with different nanometric thicknesses, were deposited by magnetron sputtering of pure elemental metallic targets on the silicon substrate. The ultrafast laser modification of the BTFs was done with an ytterbium-doped fiber laser ($\lambda = 515$ nm, $\tau = 300$ fs). Irradiations were done in the air with low pulse energies whose maximal value was 1.2 µJ. After the irradiations, the samples were analyzed by optical microscopy, scanning electron microscopy (SEM), and profilometry.

Registered morphological changes depend, for fixed wavelength and pulse duration, on other irradiation conditions (repetition rate, pulse number, static or movable target, etc.). Also, they depend on the composition of the target, as well as on the metal covering the surface of the BTFs. It is known that UFL irradiation of metals is proceeding via multiple stages as: 1) absorption of photon energy including photo excitation of electrons, 2) energy transfer from electrons to the lattice, 3) melting and thermal expansion, 4) ablation, and in some cases, lift-off a surface layer – spallation, and 5) thermal relaxation [1,2]. Cases 1 and 2 highly depend on the specific metal zone structure and different electronic dynamics of components of BTFs and are the most important at low laser fluences [3,4] as it is the case in our experiments.

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Theory for the laser induced transition from nonthermal to incoherent thermal lattice dynamics in condensed matter

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In laser excited solids, the produced hot electrons are usually described in the canonical ensemble in terms of the Helmholtz free energy, which in turns determines the potential energy surface (PES) for the ions $U(T_e) = E_e - T_e S_e$, where T_e, E_e and S_e are the electronic temperature, total energy and entropy after laser excitation and thermalization, respectively. On the other hand, the motion of the ions on the laser excited PES fulfills energy conservation as long as T_e does not change and can be treated in the microcanonical ensemble. Now, when the electron-phonon coupling is active and there is energy exchange in both directions, the mixed canonical-microcanonical usually leads to inconsistencies and wrong results. So far, different approximate approaches have been used to overcome this problem, but a microscopic theory is still lacking.

In this talk, we present a theory for the coupled description of the laser induced ultrafast dynamics of the electronic temperature and the ions from first principles. The theory yields and generalizes the equations of the two-temperature-model-molecular-dynamics (TTM-MD) method in a natural way and can be used either in ab-initio simulations or in large-scale MD simulations based on T_e-dependent force fields.

We apply this theory to study the ultrafast lattice dynamics in graphene, amorphous carbon and graphite performing ab-initio simulations and the dynamics of Sb, Si and Carbon using large-scale MD simulations based on force fields derived using our recently proposed self-learning algorithm [1]. Remarkably good agreement with experiments is obtained. The importance of using T_e-dependent force fields in MD simulations is discussed.

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Two-dimensional material printing via blister-based laser induced forward transfer

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Laser-induced forward-transfer (LIFT) is a method of printing a material from a donor to a receiver. It has been proven that high resolution and speed can be achieved for gels and liquids as well as suspended solids. The LIFT method utilises a pulsed laser to irradiate a sample of donor material and eject a small voxel of it to another surface at a defined location. In order to protect the print material from damage from the irradiation source, dynamic release layers (DRL) can be used to absorb the laser pulse energy. Utilisation of a titanium DRL, which when irradiated forms a small blister/protuberance, results in mechanical ejection of the material for printing. The work presented demonstrates the use of this technique in application to a variety of two dimensional nanomaterials, covering nano-insulators, conductors and semi-conductors. This presentation demonstrates the method and effectiveness of blister based LIFT via short and ultrashort laser pulses for the deposition of MoS_2 $MoSe_2$, hBN, and graphene flakes. The research on this avenue is rapidly growing with recent publications demonstrating similar yet distinct and yet to be refined methods for transfer of 2D graphene with a triazene DRL. [1]

With the initial graphene-production method awarded the Nobel prize in 2010 and the largest EU supported research announced in 2013 for the graphene flagship project, 2D materials beyond graphene have become a rapidly expanding set of materials to explore. The variety of single-atom-thick layered materials presents new construction medium for nanostructures and their resultant technologies. Recent focus has shifted towards stacked heterostructures [2], which enable development of complex opto-electronic and nano-electronic devices, such as sensors, transistors, waveguides and quantum computing.

We have demonstrated the efficacy of this technique in relation to direct write printing of nano-semiconductors, MoS_2, $MoSe_2$, nano-conductors, graphene, and insulators, hBN, through the use of Atomic force microscopy and Raman spectroscopy. [3] This analysis demonstrates limited damage of the materials due to the transfer process, with this technique, which enables the possibility of 2D material heterostructure generation, amongst other 2D nanotechnologies.

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Two-photon pumped polariton condensation

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Terahertz (THz) radiation sources for applications in communications, biosensing, security and others have attracted considerable interest in recent years, however, to date, their efficiency remains limited. Recently, it has been proposed that considerably more efficient THz emission can be achieved in a strongly-coupled system of exciton-polaritons in the polariton lasing regime, utilizing the effect of bosonic final state stimulation [1]. These part-light part-matter quasiparticles realize a new generation of inversionless bosonic lasers [2]. The proposed scheme relies on two-photon absorption (TPA) into the optically "dark" 2p exciton state in the cavity-embedded quantum well, such that a subsequent THz transition to the 1s-exciton-based lower polariton (LP) ground state becomes stimulated when feeding a polariton condensate. Several groups have demonstrated TPA-based excitation of non-condensed polaritons [3,4,5], yet condensation via TPA was not achieved so far due to the challenging conditions required: on one hand, extremely high intensities injecting high carrier densities by TPA, while on the other, low pulse repetition rates allowing cool down in-between pulses, all while maintaining strong light-matter coupling in a condensate-supporting structure.

Here we demonstrate two-photon pumped polariton condensation, achieved by TPA-based excitation with suitable conditions. We show this in a planar GaAs-based microcavity, by pumping at energies near half that of the lower exciton levels with a pulse spectral width covering the 2p exciton resonance. The resulting angle- and time-resolved photoluminescence (PL) (Fig. 1a-c, h respectively) exhibits a clear threshold as a function of increased TPA intensity, coinciding with an interaction-induced blueshift and a spectral linewidth narrowing (Fig. 1d-f), characteristic of the transition from a polariton thermal distribution to polariton condensation. TPA is evidenced in the quadratic dependence of the input-output curves below and above the threshold region for two detunings (Fig. 1g), and second-harmonic generation is ruled out by the threshold behaviour itself and by scanning the pump energy and observing independence of the emission peak energy. Our results pave the way for realization of a polariton-based THz laser source and shed new light on multiphoton-excited condensate formation

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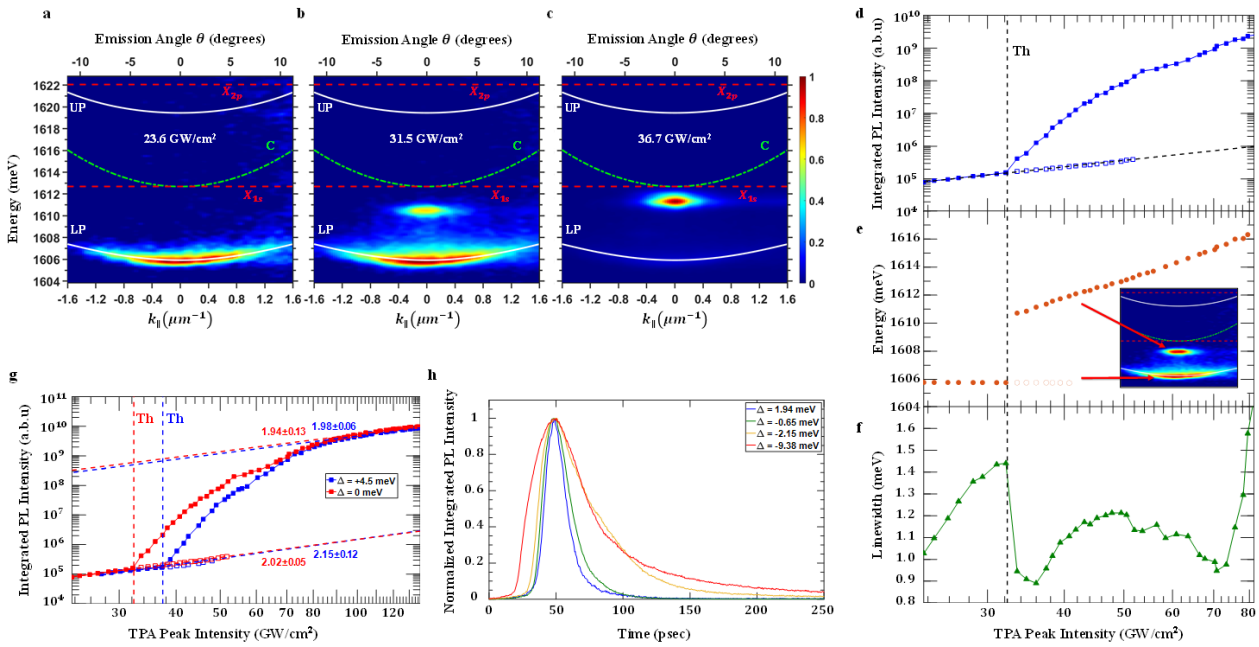

Figure 1: a-c, Angle-resolved PL at zero detuning and T=6K for three TPA peak intensities. Lines are calculated LP & UP (white solid), cavity photon C (green dashed-dotted), and 1s (X_{1s}) and 2p (X_2p) exciton (red dashed) dispersions. A transition from sub-threshold thermally-distributed LPs to a blueshifted condensate is observed. d-f, Extracted PL characteristics vs. TPA peak intensity. A clear threshold is observed at ~ 32.3 GW/cm^2 across all panels. The discontinuous jump of the emission peak (b) is due to the pulsed excitation, and the interaction-induced blueshift and spectral narrowing of this peak (c) are a clear manifestation of condensation. g, Integrated PL vs. TPA peak intensity for two detunings (red is 0 detuning, blue is +4.5meV). Diagonal dashed lines are power law fits showing good agreement with a power-of-two dependence, as expected for TPA. h, Normalized integrated condensate PL vs. Time for different detunings, showing a clear dependence of the condensate emission pulse on the LP exciton-fraction.

2D-THz spectroscopy investigation of electron dynamics in InSb and perspectives in Weyl semimetals

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Over the past decade, 2D spectroscopy has been extended to the THz frequency range, from 1 THz to 25 THz, showing the potential of this technique for probing and exciting low-energy excitations in condensed matter systems, including electronic and lattice coupling [1], 2-phonon coherence [2], magnon nonlinearities [3] and phonon nonlinearities [4].

Here, we present 2D-THz experiments in the range between 1-10 THz in a reflective geometry, investigating the electronic band-nonlinearities of a low bandgap semiconductor InSb, using THz electric fields generated from a 2-colour plasma source (up to 100 kV/cm) and optical rectification in an organic crystal (up to 600 kV/cm). In the very first picoseconds after excitation, coherent motion dominates the nonlinear response. Subsequently, electron-electron scattering effects, i.e. impact ionization, start to dominate.

Using 2D THz spectroscopy, we show that we can follow the continuous ballistic trajectory of the out-of-equilibrium electron population in the ($\Gamma \rightarrow X$, K)-plane of InSb (Fig. 1). We demonstrate that the nonlinear observations result from ballistic transport of electrons along an anharmonic and anisotropic conduction band. The spectra and their symmetries contain information regarding the local band curvature so that we can identify the symmetry of the valley in which the carriers reside.

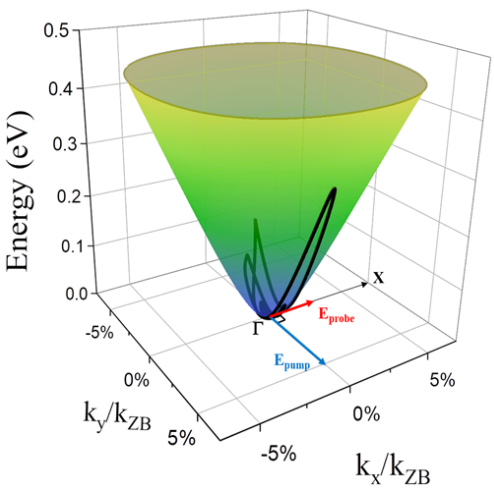

Figure 1: THz-driven electron ballistic trajectories in InSb.

This sensitive probing technique is of particular interest to investigate the low-energy linear conduction band in Weyl semimetals as well as their low-energy carrier dynamics. Preliminary results of 2D-THz investigation of THz-induced carrier acceleration in TaAs will be presented.

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Dynamics and efficiency of ultrafast laser ablation of metals and implications for MHz- and GHz-burst processing

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A surface irradiated with an ultrashort pulse passes through a sequence of physical processes, occurring on a temporal range spanning from femtoseconds to microseconds. Open questions remain as to why the pulse duration and temporal pulse separation influence the energy specific ablation volume.

Pump-probe ellipsometry (PPE) [1] reveals changes to the complex refractive index for the first tens of picoseconds, while pump-probe microscopy (PPM) [2] gives access to changes of the relative reflectivity from the initial pulse impact to the final state after about 1 µs. Fig. 1 displays PPM experiments on Cu, Al and AISI304 in air, showing the transient evolution of the relative reflectivity change $\Delta R/R$ for probe pulse delay times ranging between -12.5 ps and 10 µs.

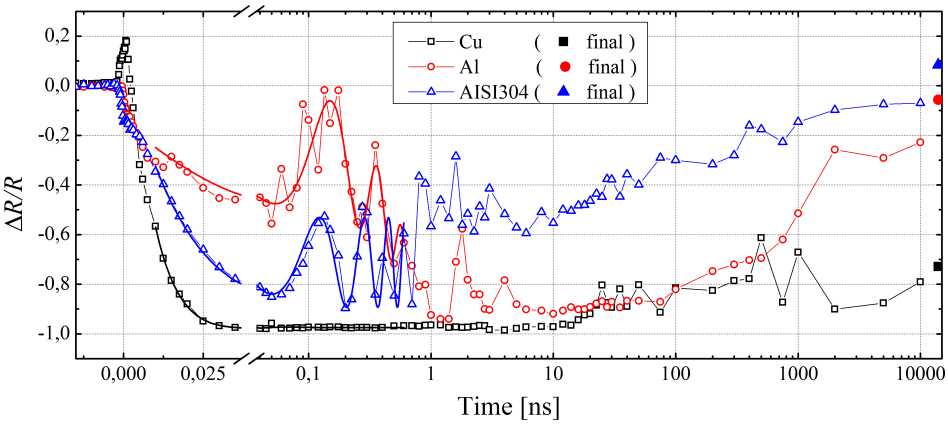

Figure 1: Transient reflectivity change ΔR/R of Cu (black open squares), Al (red open circles) and AISI304 (blue open triangles).

The experiments indicate an ultrafast density decrease at the surface of 30% within the first 5 ps, which creates a surface expansion [3] as well as separation and propagation of a spallation layer from about 100 ps to 1 ns. Finally, the spallation layer disintegration and particle generation occurs from 1 ns to 100 ns [4]. .

The ablation process is efficient and precise when it fulfills stress-confinement. This observation is valid for all studied metals, AISI304, Al and Cu. AISI304 may show a higher degree of stress-confinement due to its fast electron-phonon interaction resulting in a clean photo-mechanic spallation. Al and especially Cu display an attenuated stress confinement due to longer electron-photon interaction leading to a more photo-thermal phase-explosion, which is attributed to a higher vapor contribution in the ablation process.

We measure the energetics of single pulse ablation by determining the actual absorbed energy per volume ablated (EVA). The EVA values lie close to the total evaporation enthalpy of about 50 J/mm^3, for which Al (35 J/mm^3) lies below and for AISI304 (176 J/mm^3) and Cu (133 J/mm^3) lie above [5]. When the pulse duration increases beyond the mechanical expansion time of about 2 to 5 ps, the EVA increases by a factor of about two, observed here for single pulses. The energetics is more reflecting the mechanical properties than the optical properties of the different metals, indicating the strong photo-mechanic character of ultrafast laser ablation.

Our measurements and simulations support the conclusion that ultrafast laser ablation is efficient and precise when initiated with pulse durations shorter than the mechanical relaxation time of about 5 ps and left un-interrupted until the final state approaches after about 10 ns to 100 ns [6]. MHz-burst processing leads to particle shielding, but may be useful to split higher pulse energies into more efficient pulses fluences, when inter-pulse spacing is higher than 10 ns to 100 ns. GHz-burst processing is leading to strong shielding and even re-deposition, but the ablation from a liquid surface due to heat accumulation may be interpreted as ablation cooling.

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Molecular dynamics simulation of structural evolution in crystal and amorphous alloys under ultrafast laser irradiation

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Surface structuring of both amorphous and crystals materials may find interest in many leading technological applications. The one we target is related to laser-driven surface topography modification, however, so far, no exhaustive study was made to really give a clear picture of the different mechanisms involved in this complex process. The main originality of this work is to investigate the ultrafast phenomena and the subsequent structural modifications triggered by femtosecond laser pulses (< 100 fs). In this context, classical molecular dynamics approach is proposed. The challenge is to capture solide-solide phase transformations in both amorphous and crystal matrices. To manage the previous investigations, we have used LAMMPS, a powerful tool to lead atomistic simulation, which is adapted to catch phenomena occurring at a picosecond scale. To model this laser-mater interaction a suitable hybrid simulation combining Molecular Dynamics and Two Temperature Model is used to predict the involved dynamics of the electrons and ions. At the same time this allows us the control of laser's most important processing parameters, namely the fluence and pulse duration. We have to highlight that this work deals with metallic and amorphous based alloys structures, the latter were obtained by quenching the liquid at 10^{10} K/S. Dealing with such structures is motivated by two reasons, the experimental evidence that this alloys compositions are stable and the availability of a robust Embedded Atomic Method potential which faithfully reproduces the properties required to determine the thermodynamic path for laser-induced polymorphic phase transition.

Laser action on complicated targets: laminates and surface with a bubble

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Results obtained in the papers [1] and [2] are discussed.

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Infrared and terahertz photoconductivity of sulfur-hyperdoped Si structure

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Silicon-based mid-far IR and THz sensitive photodetectors are very interesting due to many advantages of Si as a material. In this work a photoconductivity spectra of sulfur-hyperdoped Si structure is shown (Fig. 1.). Si structure consist sub-micron thick surface sulfur-based n-doping layer on the p-doped Si wafer. It was made by femtosecond-laser induced sulfur-hyperdoping of crystalline Si wafer. Measurements were made at helium temperatures (5–105 K) and demonstrate impurity-based near-far IR (2-21 micrometers) photoconductivity spectra in the form of well-resolved separate bands of neutral and ionized atomic-like and cluster-like sulfur centers.

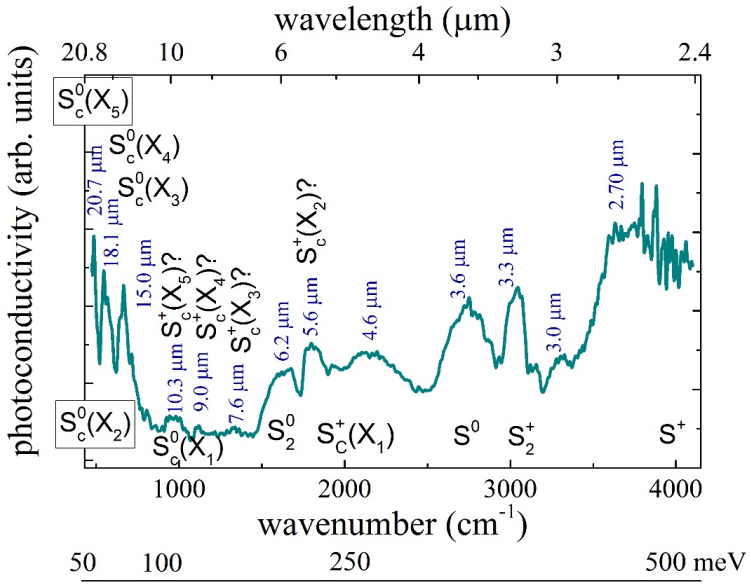

Figure 1: Photoconductivity spectra of sulfur-hyperdoped Si structure.

This study is funded by the grant of Russian Science Foundation (project no. 18-29-20022).

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Tuning the spectrum of magnetostatic waves optically excited in ferromagnetic anisotropic films

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Spintronics is an alternative to electronics, where the spin of elementary particles rather than their charge is used to code the information [1]. Spintronics is promising in solving a series of problems inherent in traditional electronics, such as high ohmic energy loss. Regarding this, a number of phenomena is of special interest, where the angular momentum transport is not accompanied by a charge current. Therefore, it is not surprising that spin waves (SWs), serving as a typical example of such transport, are currently an object of intensive study in a sub-field of spintronics – magnonics [2]. Recently, all-optical methods of SWs excitation and detection have been demonstrated experimentally [3]. On the other hand, active optical control of SWs propagation is up-to-date task in magnonics [4]. However, the rates of the control are far from the ultrafast regime yet. Therefore, exploiting femtosecond laser pulses in reconfigurable magnonics is modern challenge for fundamental magnetism with potential impact on future data processing applications.

In the presented study, we use two-color optical pump-probe technique with spatial scanning to study the influence of femtosecond laser pulses on propagation of magnetostatic surface waves (MSSW) in ferromagnetic metallic films of iron and galfenol ($Fe_{0.81}Ga_{0.19}$). The films exhibit the pronounced in-plane magnetic anisotropy. We show that the feature provides the opportunity to excite MSSW via ultrafast thermal magnetocrystalline anisotropy changes [5]. Next, we demonstrate experimentally the narrowing of the spectrum of the laser-excited MSSW wave packet as it propagates away from the excitation area [6]. Moreover, we control whether the low- or high-frequency part of the spin wave spectrum is suppressed upon propagation by changing the orientation of external magnetic field with respect to the anisotropy axes. The theoretical description of the effect is given in terms of the spatial gradient of the magnetization and anisotropy parameters of the film induced by the laser pulse. The concept of MSSW control by fs laser pulses is extended further by analysing properties of the MSSW optically excited near a Néel domain wall in a ferromagnetic strip. Micromagnetic modelling reveals the appearance of controllable resonance peaks in the MSSW spectrum and shows that the combination of femtosecond optical excitation with magnetic nonuniformity of the film, e.g. a domain wall, serves as a tuneable source of MSSW wavepackets [7].

The work is supported by RFBR (project 20-32-70149) and BASIS foundation (project 19-1-3-42-1).

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Morphological and phase modifications in amorphous $Ge_2Sb_2Te_5$ thin films on dielectric substrates driven under femtosecond laser illumination

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Chalcogenide semiconductor glasses such as $Ge_2Sb_2Te_5$ (GST225) are widely used as a basic material for rewritable and non-volatile memory applications, as well as reconfigurable nanophotonics [1]. Femtosecond laser irradiation of amorphous GST225 thin films on conductive substrates emerges both to reversible phase switching and formation of surface gratings with wavelength period [2]. However, the similar experiments weren't carried out with the samples on dielectric substrates.

In this work amorphous GST225 thin films with thicknesses 130 nm were deposited by direct current magnetron sputtering on oxidized silicon SiO_2 substrate. Femtosecond laser treatment (1250 nm, 135 fs, 10 Hz, 0.1 J/cm^2) was used for modification at the scanning mode (laser pulses per spot N_s from 3 to 750).

Scanning electron (SEM) and atomic-force microscopies (AFM) data revealed formation of ripples with period 1200-1300 nm since N_s=150. Furthermore, these grain ridges are directed orthogonally to laser polarization. Such structural properties may indicate surface plasmon-polariton generation during intensive photoexcitation of free charge carriers [3]. Sipe-Drude theory calculations suggest with this proposal for used radiation parameters [4]. For the pulse numbers N_s=300 and N_s=750 another type of ripples was observed. The elongated clusters posses subwavelength period 120-140 nm and direction which is directed perpendicularly to laser polarization. Formation of such gratings are commonly explained by combination between incident radiation scattering and thermo-mechanical effects [5]. Furthermore, Raman spectra show driven particular crystallization of the irradiated areas for the N_s=3 to N_s=15 [2]. For the greater pulse numbers (N_s=300 and N_s=750) re-amorphization of previously crystallized area was observed. Most likely, this behaviour indicates reversible phase transitions.

The research was supported by RFBR (grant № 20-32-90111).

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Laser-induced heating and precession of magnetization in Fe_3O_4 near phase transitions temperatures

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Studies of efficient approaches for changing the magnetic state of matter at ultrafast time scales has become popular rapidly developing area of condensed matter physics [1]. Among such approaches are the laser-driven a spin-reorientation transitions (SRT). SRT is characterized by a change in the direction of easy magnetization axis relative to the crystallographic axes under an external stimulus, such as temperature, pressure and magnetic field. Importantly, several works have shown that SRT can be induced by a femtosecond laser pulse in dielectric orthoferrites [4], where its mechanism is well understood by now. Recent observation of laser-driven SRT in magnetite [2], in turn, requires further elaboration.

The spin-reorientation transition in magnetite is known to occur at a temperature of 130 K [4]. Below this temperature, the magnetite possesses uniaxial anisotropy with the direction of magnetization along the c axis, and above this temperature, it has cubic anisotropy with the direction of magnetization along one of the cube diagonals. At difference with orthoferrites, in magnetite, just 7 K below SRT, the Verwey transition occurs [5]. The Verwey transition is a first-order insulator-to-metal transition (IMT), in which magnetite transforms from a dielectric phase into a metallic. Ultrafast IMT in magnetite has been studied in details in [6], while mechanism of laser-driven SRT [2] remains unclear.

In this work, we investigated the ultrafast heating induced in bulk magnetite as a result of the excitation with a femtosecond laser pulse. Our task was to determine whether the laser-induced SRT in magnetite reported in [2] can be ascribed to laser-induced heating only, or more complex electronic dynamics is involved in analogy with the ultrafast IMT [6]. For this, the temperature increase in magnetite following excitation with a femtosecond laser pulse was calculated for the range of laser fluencies 0.2-8.5 mJ/cm^2 and initial sample temperatures 80 K. As a result, it was shown that the ultrafast SRT can be reliably ascribed to laser-induced heating across the SRT temperature. In order to examine a possibility of non-thermal electronically-driven SRT in magnetite it is necessary to use lower laser pulse fluencies.

We have also evaluated expected frequencies of magnetization precession occurring as a result of laser-driven SRT. It is found that the precession frequency should be of 6–8 GHz in a field of 300 mT for the laser pulse fluencies 3 mJ/cm^2, that was observed in experiments [2].

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Structural modifications in $Ge_2Sb_2Te_5$ thin films during thermal crystallization

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The thin films of $Ge_2Sb_2Te_5$ (GST) have been successfully used as an active functional layer in the elements of nonvolatile optical phase change memory, in particular, devices of nanophotonic and integrated optic. It is possible due to the high-rate phase transformations (¡ 50 ns) from amorphous to crystalline states accompanied by the large optical contrast between states [1]. However, the lack of detailed understanding of the structural transformation mechanisms in GST thin films prevents further optimization and development of the multilevel PCM technology. So, the aim of this work is an "in-situ" investigation of the local structural rearrangements in GST thin films during thermal crystallization.

The amorphous $Ge_2Sb_2Te_5$ thin films (30nm thickness) were deposited by the magnetron sputtering on the Si and SiO_2 substrates with planar Al electrodes. The optical properties in the range from 300 to 2100 nm were measured by ellipsometry. The temperature dependences of the Raman spectra and resistivity for the GST thin films were obtained through simultaneous measurements in the temperature range from 30 to 350°C in an Ar atmosphere. The seven peaks in the obtained Raman spectra were decomposed by the Gaussian fitting. The results of the investigation of GeTe and Sb_2Te_3 bulk materials were used for the association of the peaks with structural unit vibrations.

Analysis of the Raman spectra temperature dependences showed that heating leads to changes in the peak parameters (intensities, peak areas, FWHMs and shifts peak position). These changes occur in the temperature ranges of 145 − 160°C and 265 − 290°C and can correspond to the transitions from an amorphous state to fcc, and further from fcc to hcp phases. It should be noted that the sharp changes of the refractive index were also observed in the identified temperature ranges of phase transition. All of these property modifications in total indicate the ongoing structural rearrangements.

A quantitative and qualitative analysis of the temperature dependences of the Raman spectra showed that the crystallization of GST from the amorphous state to the metastable fcc state takes place through the rearrangement of Ge/Sb atoms from tetrahedral and pyramidal configurations into defective octahedral one for GeTe and Sb_2Te_3 compounds, respectively, whereas the fcc→hcp transition takes place primarily due to the local rearrangements of Sb atoms caused by the elongation of the covalent bonds.

This work was supported by Russian Science Foundation (project № 20-79-10322).

Numerical investigation of chirped laser pulse focusing in bulk of Si

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The problem of achieving extreme laser fields and the density of free electrons with the aim of microstructuring bulk semiconductors is of current interest [1-3]. The possibility of creating bulk micromodifications inside Si using picosecond laser pulses with a wavelength of 1.5 μm was recently demonstrated [4]. Some restrictions on the duration of laser pulses and the numerical aperture of the beam were formulated, which make it possible to reduce the nonlinear absorption of laser radiation in the prefocal region and ensure the delivery of the energy required for microstructuring to the focal region. Such picosecond pulses are not spectrally limited, and the phase difference between spectral components can affect pulse propagation.

In this paper, we numerically investigate the role of a pulse chirp in overcoming the nonlinear delocalization of laser energy in the bulk of Si. In the calculations, we used laser pulses with a wavelength of 1.24 mm and a duration from 120 fs (spectrally limited) to 400 fs (chirped both positively and negatively) with an energy of 1 µJ.

To simulate the propagation of laser pulses in the bulk of Si, we used the unidirectional pulse propagation equation (UPPE) [5]. Photoionization is described by the Keldysh formula and, together with avalanche ionization, is included in a multilevel ionization model [6], which allows one to calculate the dynamics of the density of free electrons.

Calculations show a non-monotonic dependence of nonlinear absorption on the pulse chirp. The maximum value of 0.90 is achieved with a positive chirp of 10000 fs^2, which corresponds to a pulse duration of about 150 fs. A decrease in chirp results in an approximately directly proportional decrease in nonlinear absorption. In contrast, for a positive chirp exceeding the optimal value, the absorbed pulse energy decreases faster. This behavior is associated with a strong delocalization of the laser energy in the prefocal region. For longer pulses, the avalanche ionization mechanism prevails, which, as calculations show, leads to the formation of a denser electron plasma. This leads to strong nonlinear absorption in the prefocal region. The shift of the maximum of the absorbed pulse energy towards the positive chirp can be explained by a decrease in the phase difference between different spectral components due to the combined action of the Kerr and plasma nonlinearities. It is also shown that when the medium is irradiated with a pair of pulses separated in time, the nonlinear absorption of the second pulse increases to 0.94.

This research was partially supported by the Russian Foundation for Basic Research (Project № 21-32-70021 and 18-02-40014). K. V. Lvov is a scholar of the Foundation for the Advancement of Theoretical Physics "BASIS".

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Ultrafast optical modulation in dielectric nanoantennas and Weyl semimetal thin films

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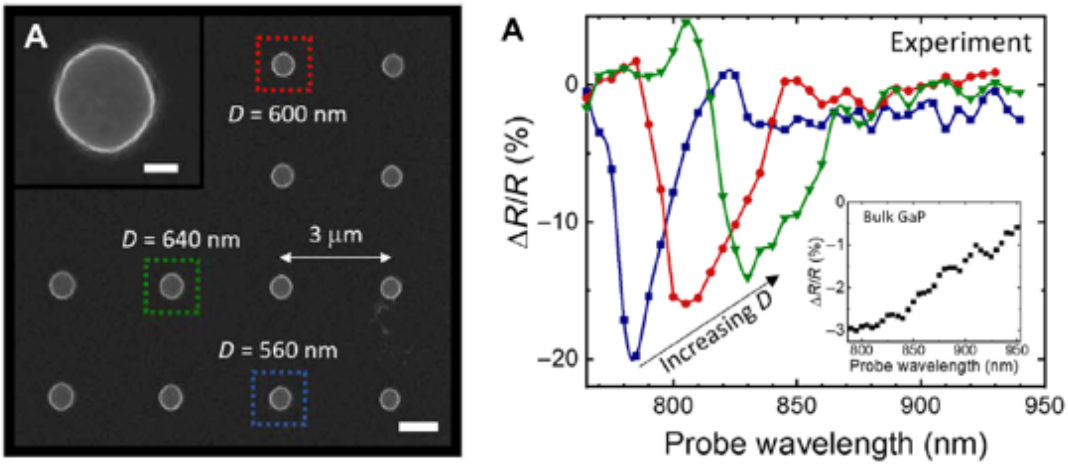
Metallic and dielectric nanostructures provide distinct and unique means for shaping the electromagnetic near field, and for channelling radiation from the far field to the nanoscale. The associated electromagnetic field hot spots can be exploited for the enhancement of interactions between light and matter, most prominently for surface-enhanced spectroscopy and sensing, the boosting of non-linear interactions, and also for nanoscale spatial control over chemical reactions.

In my lecture I will present an overview of our work on utilizing dielectric nanoantennas consisting of GaP for ultrafast fs-scale optical modulation with record modulation depth. I will also present initial results on sub-fs all-optical modulation in Weyl semimetal thin films.



Reflectivity modulations of a single GaP nanodisk excited at its anapole excitation frequency (Science Advances 6:eabb3123 2020).

Time-resolved diagnostics of laser-induced extreme state of condensed matter and supercritical fluids

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We offer an approach that combines time resolved shadow photography and Raman spectroscopy for study of the laser-induced extreme state of matter. We used tightly focused ultrashort laser pulse for plasma generation. The energy conserved in the electron subsystem during plasma relaxation leads to the shock wave generation, cavitation bubble formation and phase transitions (including phase transition to supercritical state). In experiments, two laser systems (1054 and 527 nm, pulse duration is 6 ns with frequency up to 4 kHz and energy up to 1 mJ) were synchronized with a 250 ps time resolution. The IR laser pulse acted as a pump and visible laser acted as a probe (for both shadow photography and Raman spectroscopy). The time delay was varied automatically (in the range from 250 ps to ms). Using Raman spectroscopy, we obtained the evolution of the vibrational spectra of CO_2 and water. There red shift and spectral broadening of the Raman-active lines indicates the transition to supercritical state [1]. The broadening is changed with temperature and is maximal at the so-called Widom delta (see Fig. 1). Using shadow photography we retrieved the pressures induced by the pump pulse [2]. Thereby we retrieved the pressure and temperature of the laser-induced extreme state of matter.

The work is supported by RFBR grants 19-32-60072, 18-29-06056, 18-29-0603.

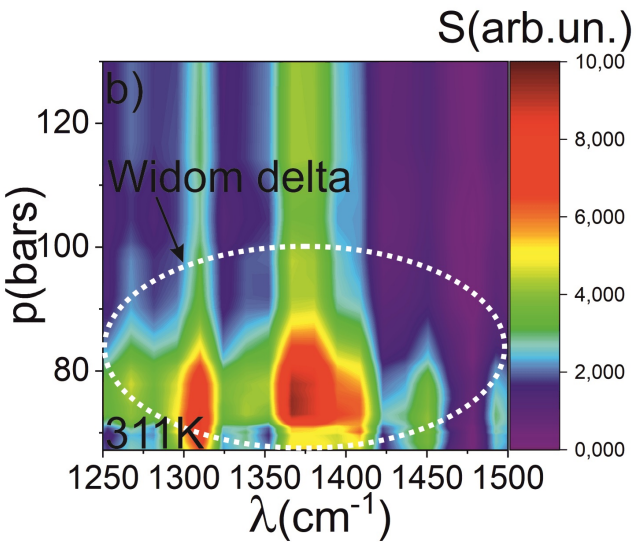

Figure 1: The Raman spectrum of sc-CO_2 for different pressures under temperature of 311 K.

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Interaction of tightly focused mid-IR femtosecond laser pulses with Si

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We demonstrate a comparison of the effect of pulses with different durations (from 0.1−0.2 ps to 1.2 ps) of the near (1.24 µm) and middle (4.6 µm) infrared under tight (NA = 0.85) focusing of laser radiation into the bulk of the silicon (Si). In the mid-IR region, the ionization mechanism is within the framework of the multiphoton approximation. However, an increase in the photon order for excitation by mid-IR pulses leads to a decrease in delocalization processes and losses in the pre-focal region [1]. The development of high-power mid-IR laser applications requires a study on laser-induced damage threshold in the mid-IR. We demonstrated that in semiconductors (for example, Si), greater absorption (80 %) is achieved for longer (sub-ps) laser pulses [2]. Under such conditions, it is possible to create micromodifications inside the bulk of the material as the deposited energy density is higher than the threshold value. These results will serve as the basis for the development of modern microprocessing technologies since the creation of modifications in the bulk of a semiconductor in this case does not require complex schemes of solid-state immersion or space-time chirping.

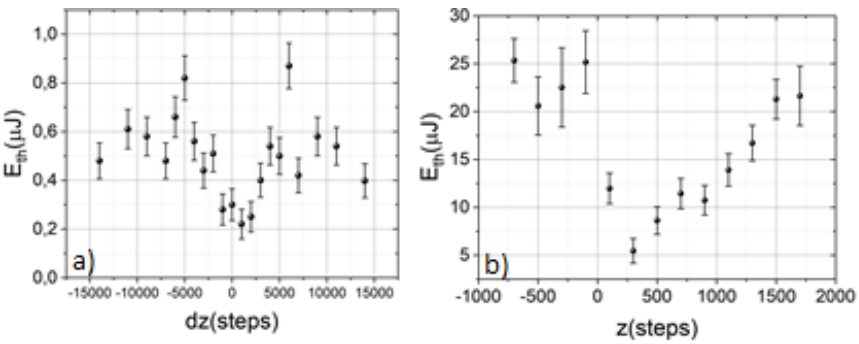

Figure 1: Plasma formation threshold as a function of the detuning length of the femtosecond compressor for different solids and pulse durations a) Si 1.24 µm, b) Si 4.6 µm.

This work was supported by Russian Science Foundation (RSF) (Project No. 17-72-20130) and partially by the Russian Foundation for Basic Research (Project No. 21-32-70021 and 18-02-40018).

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Hot electron ultrafast termalization in high-Tc cuprates

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Because of the large value of superconducting gap in cuprates a transient non-thermal population can be created by ultrashort and very intense THz and far-infrared (FIR) pulses (see, e.g. [1]). Hot electron system 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. Optical excitation of complex materials with ultra-short FIR and MIR laser pulses allows to induce and dynamically control non-equilibrium unconventional orders (e.g., AF SDW). Generation of CT-excitons in the normal state of high-Tc cuprates can unleash superconductivity far above the equilibrium critical temperature [2,3]. A light-enhanced drop of the sample resistivity can be substantiated by direct electrical transport measurements performed under the same excitation conditions. This confirmed the long timescales and revealed a light-enhanced state with vanishing resistance occurring in the normal state. Such substantial positive photoconductivity being unusual for a metal, and in combination with the transient optical conductivities, it is suggestive of metastable light-enhanced superconductivity [3]. In this work, the detailed analysis of available ultrafast optical data is performed on the basis of an electronic pseudotemperature concept [4]. The results demonstrate that the entire landscape of relaxations can be understood by modeling the system as following a nonequilibrium, electronic pseudotemperature that controls all electrons in the zone. The approximate time after which the hot electrons have thermalized with each other (but not necessarily with phonons) in cuprates is about 200 fs while thermal decay time for the hot electron population there appears to be near 4 ps. In the superconducting state, the thermal response is well described by a single exponential decay, at least out to 12 ps. After the electronic thermalization time scale, the parameters relevant to superconductivity, energy gap Δ and scattering rate Γ , follow parametrically from the electronic pseudotemperature. This simplifies the interpretation of quasiparticle population dynamics in the postthermalization regime and gives a coherent picture of the decay dynamics in photoexcited cuprates.

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Phonon driven ultrafast nonlinear dynamics in a Bi_2Se_3 crystal

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Selective excitation of coherent high-amplitude vibrations of atoms in a solid can induce exotic nonequilibrium states, in which the character of interactions between electronic, magnetic and lattice degrees of freedom is considerably altered and the underlying symmetries are broken. We use powerful single-cycle THz pulses to pump an infrared-active phonon mode of a Bi_2Se_3 crystal. The resulting intense coherent atomic vibrations induce a short-lived nonlinear lattice distortion, which reveals itself as dynamical splitting of the doubly degenerate Raman-active phonon mode. We interpret this distortion as the result of excitation of anisotropic electronic distribution via quadratic coupling to the THz-driven phonon mode. The observed effect represents an example of the ultrafast phase transition in a topological insulator and bears similarity to the physics of electronic nematic phases. We also observe nonlinear generation of coherent Raman-active phonons of four symmetries and discuss its possible mechanisms: lattice anharmonicity and sum-frequency Raman scattering.

Thermal and non-thermal phase transitions in laser-irradiated metals

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When a sufficiently powerful ultrashort laser pulse interacts with a material, a phase transition to a new liquid or solid phase is possible. Various pathways from an initial to a final state can be realized depending on material type and laser irradiation parameters. In general, there are two possible scenarios. The excited hot electrons transfer their energy to a relatively cold lattice via electron-phonon coupling mechanism. Such a kinetic energy transfer may lead to a *thermal* melting of a target. Alternatively, strongly excited electronic system may weaken the interatomic potential, which results in atomic disordering without a direct transfer of thermal energy from electrons to atoms. This process is known as a *non-thermal* phase transition.

In this work, we explore the dynamics and mechanisms of thermal and non-thermal transitions in metals. We study conventional thermal melting within the two-temperature model and molecular dynamics formalisms on the example of single and polycrystalline free standing gold films. We confirm that the melting time is sensitive to electron-phonon coupling, opening a way to extract this parameter from ultrafast diffraction experiments [1] combined with theoretical analysis. Comparing the melting dynamics between single and polycrystal we conclude that the grain boundaries play a noticeable role only at relatively low absorbed fluences.

We also show with our tight binding molecular dynamics simulations that metals, in fact, should be able to undergo non-thermal phase transitions if allowed to expand due to laser-induced electronic pressure [2]. Therefore, we predict that such transitions can be experimentally detected in small nanoscale samples, such as ultra-thin films or nanoparcticles.

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Surface structuring of Ti-based implant alloy by fs laser light – low and high fluence mode

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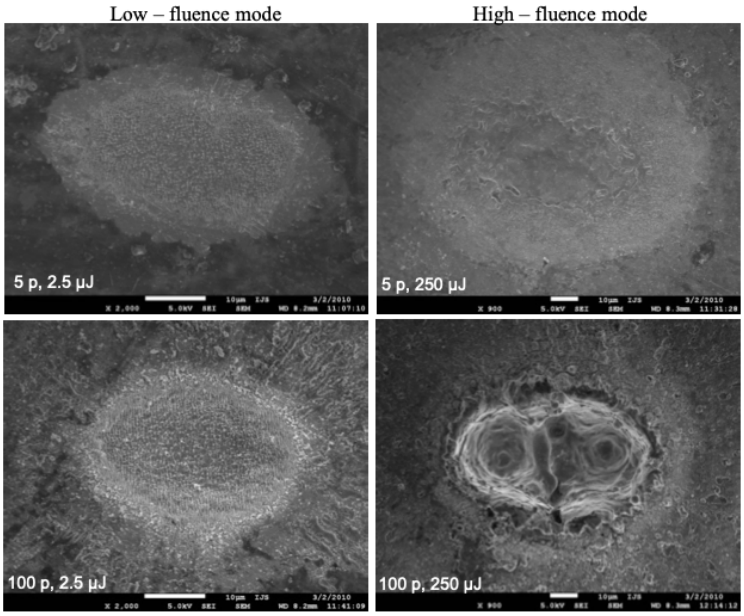
Research of surface modifications of the material by laser beams, especially with ultra-short pulses, is a field of research with significant impact on its application in nano- technology and bio-mechanics. Ti6Al4V alloy is one of the mostly used titanium alloys in biomedical applications due to its excellent combination of mechanical properties and chemical stability[1]. Therefore, it is of great interest to adeqautely prepare or modify the alloy surface for specific applications, for example, adjusting the Ti6Al4V surface for osteoblast growth if the alloy is used as a bone implant. During the presented investigations, the femtosecond Ti:sapphire laser (Clark CPA-2101) was applied for Ti6Al4V surface modification. Experiment was conducted in air by a focused laser beam with 775 nm wavelength and pulse duration of 200 fs. The surface of the sample was irradiated with the count of pulses varying from single pulse and up to 100 pulses, with increament of the energy density from 0.04 J cm^{-2} to 4.55 J cm^{-2}. Laser action induced ablation of the surface material and two important parameters as the laser ablation threshold fluence (F_{th}) and the incubation factor (ζ) of the material were calculated. Besides ablation and hydrodinamic effects, the appearance of laser induced periodical nano-structures and micro-/nano- particles were registered and analyzed.



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Ultrafast laser-induced phase transition in VO_2 for picosecond acoustics

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Among ultrafast phenomena in solids, photo-induced phase transitions (PIPTs) attract considerable attention as they involve sharp changes of material properties on sub-picosecond timescale. A good example is VO_2 showing a 1st-order PIPT upon femtosecond laser excitation, changing from monoclinic insulator to tetragonal metal [1]. The PIPT in VO_2 results in huge modifications of optical constants [2] as well as $\sim 0.5\%$ lattice change [3], leading to numerous applications of VO_2 [4].

Another ultrafast phenomenon is generation of coherent acoustic phonons in thin absorbing films by femtosecond laser pulses [5]. This effect lays in foundation of picosecond acostics which study generation, evolution and detection of strain pulses with amplitudes $\sim 10^{-3}$ and frequencies up to a few THz. Picosecond acoustics is used in acoustic nanoscopy [6] and as a non-thermal excitation of processes in solids [7].

In this work we combine PIPT and picosecond acoustics in two different pump-probe experiments on nanoscale VO_2 structures.

Firstly, we aim to exploit large ultrafast lattice changes during 1st-order PIPT in VO_2 to generate a picosecond strain pulse non-thermally. To achieve this we excited a 100-nm VO_2 film with femtosecond laser pulses both inducing PIPT and creating a picosecond strain pulse. Taking into account known strain generation mechanisms [8], we extracted a $+0.45\%$ non-thermal contribution from PIPT. The existence of PIPT in VO_2 allowed us to generate a high 0.7% amplitude picosecond strain with only 28 K heating of VO_2, which is three times cooler as compared to the same strain with no PIPT contribution [9].

Secondly, we used picosecond strain pulses generated elsewhere to affect the PIPT in 70 nm-high VO_2 nanohillocks. To do it we excited PIPT when the strain pulse was injected in VO_2. We found a $\sim 1\%$ change of the number of nanohillocks undergoing PIPT, which depends on average strain amplitude in a nanohillock during PIPT. Owing to bipolar nature of the strain pulse, we were able to control the amplitude and sign of this change, effectively modulating the PIPT efficiency in VO_2, by varying a time delay between the two pulses [10].

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Anisotropy of electron distribution function in metals, semimetals and graphene at subpicosecond timescales

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In this talk we summarize our latest theoretical and experimental results on the asymmetry of the electronic distribution function induced by ultrashort laser and terahertz pulses in the conductors. Special attention is paid to its manifestation in nonlinear electromagnetic phenomena and to current problems in the theoretical interpretation. In particular, the effects of non-reciprocal generation of THz pulses in monocrystalline bismuth [1], THz-driven optical luminescence of graphene [2] and the laser-induced magnetic field inside metals are compared [3]. The rate of electron momentum relaxation is critical for the electrodynamical properties of all discussed media at sub-picosecond timescale.

We also revisit our previous experimental observation of asymmetric THz response of bismuth mono- and polycrystalline samples [1]. Taking into account the possibility of low-frequency current generation by the asymmetric distribution function, we attribute anisotropy in THz response to the shifted electron distribution formed during the interband transitions between the asymmetric energy bands of bismuth [4].This allows to qualitatively explain the THz response asymmetry with respect to a half-turn of a monocrystalline sample and the THz electric field orientation relatively to the sample surface. So, here interband transitions can be treated as another source of distribution function anisotropy, not discussed in paper [3].

Another way to produce anisotropic distribution function in solids is a direct action of half-cycle THz electric field. We present our recent experimental and numerical results on the graphene excitation by strong THz pulses and analyze it, focusing on the problem of the distribution asymmetry in k-space. Spontaneously emitted photons are mostly polarized perpendicular to the THz filed, which allow us to estimate the distribution function distortion. We believe that besides the general theoretical interest to laser–matter interaction, these examples of formation and relaxation of the electronic distribution functions might be useful for interpretation of the experiments on optical-to-THz conversion, second harmonic generation, and ultrafast demagnetization of metals.

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Multifunctional IR sensor platform fabricated by direct laser printing

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We demonstrated a novel plasmonic IR sensor platform based on arrays of nanoantennas fabricated via the highly-efficient non-ablative direct laser printing on thin glass-supported Au films. The fabricated arrays of antennas, either nanobumps or nanojets, exhibited a pronounced plasmon resonance in the near-IR spectral range, which is easily tailored by tuning the deposition energy of the laser pulses or the nanoantenna arrangement [1].

Fabricated IR sensor platform was tested regarding their application for refractive index sensing, gas detecting, precise identification of the thickness of the adsorbed nanolayers [2] as well as tailoring photoluminescence of the attached IR-emitting quantum dots [3]. Remarkable sensing performance being combined with flexibility of the design, tunability of the resonances as well as facile inexpensive fabrication at extremely fast printing rate made proposed sensor platforms promising for various nanophotonic as well as bio- and chemo- sensing applications.

This work was supported by the Russian Foundation for Basic Research (Grants 20-32-70056).

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Ultrafast dissociation dynamics Induced in $(Fe(CO)_5)_nXe_m$ mixed nanoclusters by resonant femtosecond IR laser radiation

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Dissociation dynamics of $(Fe(CO)_5)_nXe_m$ mixed molecular nanoclusters induced by femtosecond IR radiation in 5 µm region are studied for the first time. The time-resolved method of resonant broadband IR excitation of CO vibrational bands in the mixed cluster's molecular core and photoionizing probing (λ = 400 nm) of its decay products (xenon atoms and iron pentacarbonyl molecules) combined with the time-of-flight (TOF) mass-spectrometry is applied.

Mixed molecular clusters were formed during supersonic expansion through the pulsed nozzle. The mixture of $Fe(CO)_5$ and Xe gases (1/300) was used at room temperature and total pressure of 200 kPa above the nozzle.

It is found that IR excited mixed clusters in the initially cold molecular beam are heated and decomposed as a result of relaxation processes, giving rise to free neutral Xe atoms and $Fe(CO)_5$ molecules. Thus formed particles are the origin of ion signal from Xe^+ and $Fe(CO)_5{}^+$ ions which grows on picosecond time scale (see fig. 1).

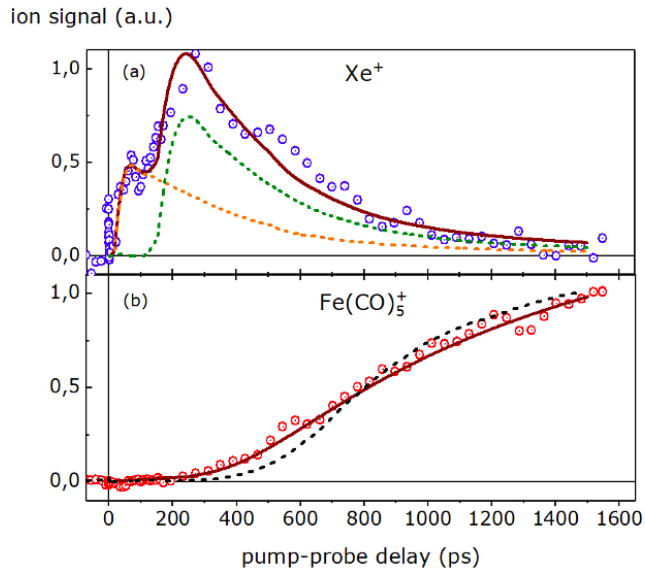

Figure 1: Normalized kinetics of the cluster's decay products: Xe atoms (a) and $Fe(CO)_5$ molecules (b). Smooth curves are the result of model calculations.

Proposed mechanism of vibrational relaxation process includes several stages. Initial laser excitation of CO vibrational bands in clusterized molecules relaxes on picosecond time scale with subsequent population of lower frequency vibrational modes of the molecules in the cluster. 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 particles according to the hierarchy of binding energies: weakly bonded shells of Xe atoms are evaporated much faster than the $Fe(CO)_5$ molecules from the cluster's core.

This work was supported by the Russian Foundation for Basic Research, grant № 20-02-00146.

Effect of the laser pulse polarization ellipticity on laser-induced acoustic wave under plasma-mediated laser excitation of the condensed medium bulk

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The effect of the laser pulse polarization ellipticity M [1] on laser-induced acoustic wave under plasma-mediated femtosecond (Cr:Forsterite laser system - 1240 nm, 100 fs, 10 Hz, energy up to 3.5 mJ) laser excitation of the condensed medium (distilled water) bulk was experimentally studied. The effects of the laser pulse energy and the degree of polarization ellipticity on the generation of an acoustic pulse are compared (Fig. 1).

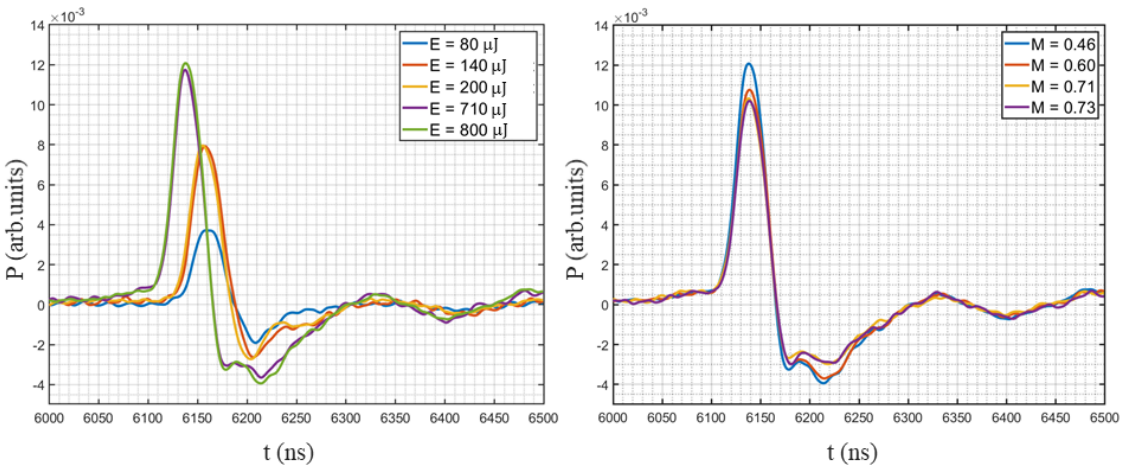

Figure 1: Left: dependence of acoustic signal on laser pulse energy E. Right: dependence of acoustic signal on laser pulse polarization ellipticity degree M.

It is established that the increase of polarization ellipticity degree leads to the decrease of the acoustic pulse energy. The maximal decrease of acoustic pulse energy is reached under circular polarization ($M = 1$) and equals about 50 % of the acoustic pulse energy under linear ($M = 0$) polarization. It is shown that this effect can be described by the theory of ionization in strong laser field [2].

The work was supported by the RSF grant № 17-72-20130. Rumiantsev B. V. is the scholar of the foundation for the advancement of the theoretical physics and mathematics "BASIS".

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Nanofabrication of composite nanostructures by femtosecond laser ablation

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Laser ablation is a versatile tool allowing manufacturing of different nanostructures without any additional chemical precursors that is an important point for various applications. Laser-based nanofabrication in liquids leads to a direct formation of colloidal solutions of single- and multi-component nanoparticles in a required liquid. In particular, a variation of experimental conditions provokes synthesis of bimetallic nanoparticles significantly tuning their plasmonic features or combining, for instance, plasmonic and magnetic properties.

However, a possibility of laser-based manufacturing of composite semiconductor/metallic nanostructures is still challenging attracting the research interest since last years. This method is shown as an easy way of combining both semiconductor and metallic elements (e.g., silicon and gold) in a form of one nanoparticle integrating different modalities [1]. It is also found that experimental conditions significantly influence physicochemical properties of synthesized nanomaterials [2]. Their promising applications, for instance, for molecule detection using surface-enhanced Raman spectroscopy via plasmonic properties of semiconductor-based nanostructures is demonstrated [3].

This research can be considered as a first step to nanofabrication of chemically ultrapure multi-component nanomaterials using ultrafast laser processing for their subsequent applications in different fields, such as nano- and biosensing, bioimaging, catalysis and photovoltaics.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 897231 (LADENTHER). Authors also thank the European Regional Development Fund and the state budget of the Czech Republic (Project BIATRI: CZ.02.1.01/0.0/0.0/15003/0000445), the Ministry of Education, Youth and Sports (Programs NPU I-Project no. LO1602).

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Laser modification of semiconductor films with Ag NPs

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Semiconductor films with plasmonic nanoparticles have a lot of applications in photonics as photosensitivity sensors, optical filters, active elements for the photovoltaics and plasmonic and other. Sol-gel TiO2 and AlZnO films with silver nanoparticles (Ag NPs) are the most demanded. The optical properties and electronic structure change and its correction for such films are widely used by laser exposure. The CW UV radiation and femtosecond pulses with high repetition rate (more than 200 kHz) are the most interesting laser sources for the films processing.

In this work the different mechanisms of laser modification, such as photothermal and photochemical structure modification was considered. We studied the modification of TiO2:Ag and AlZnO:Ag films by laser action of CW UV radiation (405 nm) and femtosecond laser pulses (250 fs) with wavelengths of 515 nm and 343 nm at a pulse repetition rate of 500 kHz and higher. The modified regions were recorded by scanning at a velocity from 0.1 to 50 mm/s and power density from 0.8 to 1.1 MW/cm^2 for CW radiation and at an fluence from 0.05 to 0.5 J/cm^2 for the femtosecond pulses.

The change in the optical constants and band gap energy of the films as a result of laser action will be studied. An explanation will be given to the mechanism of film structure modification.

The study is funded by the grant of Russian Science Foundation (project № 19-79-10208).

Electrophysical anisotropy of femtosecond laser-irradiated phosphorous- and boron-doped amorphous silicon films

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Amorphous hydrogenated silicon (a-Si:H) modified by femtosecond laser pulses is a perspective material for applications in thin-film photovoltaics and optics [1]. Femtosecond laser-induced nanocrystallization increases conductivity of the a-Si:H films and reduces their photodegradation. Simultaneously, formation of one-dimensional laser-induced periodic surface structures (LIPSS) due to plasmon-polaritons excitation is possible under the action of femtosecond laser pulses. Such structures increase optical absorption and also manifest birefringence, dichroism [2], as well as electrophysical anisotropy [3].

In this work, 400 nm thick phosphorous-doped a-Si:H (n-a-Si) and 1200 nm thick boron-doped a-Si:H (p-a-Si) films were irradiated by femtosecond laser pulses ($\lambda = 1250$ nm, $\tau = 150$ fs, $\nu = 10$ Hz). The laser fluence varied from 0.15 J/cm^2 for n-a-Si films to 0.3 J/cm^2 for thicker p-a-Si films. The laser beam scanning speed also varied from 50 to 200 µm/s.

The structural and electrophysical anisotropy of irradiated films was investigated. On the modified surface we observed formation of LIPSS orthogonal to laser polarization. Their period was close to the laser wavelength and varied from 840 ± 70 to 1100 ± 100 nm with increasing scanning speed. LIPSS relief at the same time deepened from 100 up to 400 nm. Raman spectroscopy revealed crystalline Si phase formation within irradiated films, which volume fraction decreased with film depth from $82 \pm 13\%$ to $41 \pm 6\%$ for p-a-Si, and from $19 \pm 3\%$ to $9 \pm 5\%$ for n-a-Si.

Dark conductivity of irradiated p-a-Si increased by 6 to 7 orders, up to $1.2 \cdot 10^{-2}$ S/cm due to the crystalline Si phase formation. The conductivity of irradiated p-a-Si was nonlinear, which can be caused by nonuniform distribution of crystalline Si phase volume fraction within film depth. On the other hand, the conductivity of n-a-Si did not change substantially after femtosecond laser irradiation and remained on the order of 10^{-6} S/cm. In this case the crystalline Si phase formation may be countered by excessive film ablation as the formed LIPSS relief was comparable with film thickness. Dark conductivity anisotropy emerged in all samples as a result of LIPSS formation. Its value was up to 10 times higher along the surface grooves, which may be explained by depolarization influence of the LIPSS, surface relief ablation and uneven crystalline phase distribution within a-Si:H films.

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LIPSS Regularity on Silicon: From Spot Size Effect to the Emergence of Periodic Columns of Structures

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Laser induced periodic surface structures (LIPSS) writing with femtosecond pulses is a handy approach for flexible, fast and single step nano-/micro-structuring of various materials. For different applications, it is desired to produce the LIPSS on large areas with a high level of homogeneity [1-3] and controlled periodicity [4]. In this work, we present a study of LIPSS formation on the surface of monocrystalline silicon with an emphasis on two aspects of large-area structuring.

The first aspect is the regularity of the pattern, extending previous findings [1-5] by investigating the effect of the laser spot size, a potential key to scale up high-quality surface texturing. A series of experiments have been performed by scanning the sample with linearly polarized laser pulses of 1030 nm wavelength, Gaussian spatial and temporal profiles, and duration (FWHM) of 250 fs. The regularity has been inspected using the dispersion in the LIPSS orientation angle (DLOA) [3] from scanning electron microscope images. The irradiation parameters (spot size, scanning speed, pulse energy) for reducing DLOA and, hence, enhancing regularity have been identified.

In a second part, we reveal a range of laser scanning parameters where the LIPSS appear on column-shaped areas, which are perpendicular to the scanning direction. Interestingly, the columns where the LIPSS emerge are regularly spaced and primarily not located in the regions where the fluence peaks. The formation of the LIPSS columns is examined by analyzing an intricate connection between local cumulated fluence, N-on-1 shot damage geometry and thresholds of modifications.

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Investigation of ambipolar diffusion of electron-hole plasma under the action of ultra-short laser pulses on a silicon

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Today silicon is the main material in optoelectronics, photovoltaics and electronics [1], while laser processing by ultrashort pulses is becoming more and more popular. In ultrashort pulse ablation, it is important to study the energy deposition at subpicosecond times. As is known, the main mechanism of energy deposition in semiconductors and dielectrics is due to interband generation and heating of an electron – hole plasma (EHP) with inverse relaxation dynamics proceeding through the three-particle Auger recombination of the EHP. In addition, the ambipolar diffusion of a dense EHP in semiconductors strongly depends on the EHP density [2, 3]. In this work, the coefficient of ambipolar diffusion in a dense EHP in silicon at the ablation mode is measured by measuring the radius of the craters, depending on the width of the laser pulse. It was found that the ambipolar diffusion of the plasma was stopped by Auger recombination and thermalization of the electron lattice on a time scale of ≈ 3 ps.

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Anisotropic resistivity ITO surfaces produced by laser induced self-organization mediated by fluence selective compositional modification

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Transparent conducting oxides (TCOs) are materials with low optical absorption in the visible [1], which makes them particularly suitable for key applications in information technologies and energy harvesting. Among them, Indium Tin Oxide (ITO) plays very important role, especially in niche applications where the chemical stability in water solutions is a decisive factor (electrochemical sensors).

In this work we demonstrate the production of highly anisotropic resistivity surfaces by fs-laser irradiation of ITO films at high repetition rate at 1030 nm [2]. Electrical anisotropy appears as a consequence of the formation of Laser Induced Periodic Structures (LIPSS) at the material surface through a novel mechanism associated to the dynamical modification of the material composition at the laser exposed regions. LIPSS [3] have been coherently extended over cm-sized regions and we have identified two main optimized processing conditions. At high fluence, nearly complete ablation at the valleys of the LIPSS and strong ablation at their ridges, accompanied by a preferential In-loss, lead to an insulating structure in the direction transverse to the LIPSS and conductive in the longitudinal one. At a lower fluence, the material at the LIPSS ridges remains essentially unmodified while partial ablation is observed at the valleys. The latter structures show a longitudinal conductivity twice the transverse one, and a resistivity similar to that of the pristine ITO film. The compositional changes induced as laser pulses accumulate, condition the LIPSS evolution and thus the result of the structuring process.

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Novel applications of ultrafast laser ablation in liquids

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Our group has been exploring the femtosecond (fs)/picosecond (ps) ablation technique in liquids/air for creating a variety of nanoparticles/nanostructures of Ag, Au, Ag-Au, Au-Cu, Si. We have tested their efficacy for trace explosives (ammonium nitrate, picric acid, RDX, HMX, NTO, TNT etc.) detection achieved using the Surface Enhanced Raman Spectroscopic technique [1-19]. We have successfully created flexible, robust, versatile, cost-effective, paper-based and nanofiber-based SERS targets using the nanoparticles produced from fs/ps ablation for common explosives detection using a portable Raman spectrometer paving way for point-of-interest detection. We have also extended this detection towards other hazardous materials (pesticides such as Thiram, CWA simulants). Additionally, we have explored the ablation of materials such as Si, HfO_2, GaAs [20-23] using ultrashort laser pulses resulting in novel phases. Interesting results obtained from these studies will be presented.

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Energy stability limits of precision in ultrafast laser material processing

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In laser manufacturing, precision is a key aspect when aiming at small and controllable modifications for specific applications. The size and characteristics of the produced features are defined by the delivered spatio-temporal energy distribution to the material. This makes the achievable stability and controllability of the laser conditions a critical technological question. By experiments, we study the importance of this limit and explore laser stabilization schemes to improve material processing performances.

In recent work, we established that the concept of nonlinear resolution is not applicable for femtosecond laser ablation. Consequently, there is a systematic one-to-one mapping between produced ablation features in dielectrics and beam contours at a strict threshold intensity [1]. Here we show that the same conclusion holds also for silicon surface amorphization with 200-fs pulses. This confirms an observable based on a deterministic threshold-based response and a resolution problem that is totally independent of the nonlinear confinement of absorption [1].

An important consequence of this strict threshold response is the possibility to derive the processing repeatability by using a simple 'noise' model to account for pulse-to-pulse energy fluctuations [2]. By simulating laser fluctuations in an experiment, we demonstrate the validity of the model to describe the repeatability of the produced amorphization features. In this way, we can assess the practical precision limits specific to our experiment and the stability performance of our laser.

From this, we explore concepts to exceed this limit. There are various methods to improve laser stability including dynamically controlled Pockels cell [3] or Kerr interactions [4]. Here, we propose a very simple extra-cavity method based on passive propagation through nonlinearly absorbing materials [5,6]. Using our 200-fs pulses, we measure nonlinear absorption in dielectrics (incl. Al_2O_3 and SiO_2) with band gaps from about 3 eV to 10 eV. We compare the measurements to a stabilization model assuming pure multiphoton absorption. The results show that all materials are not equally performing as some materials introduce important beam distortions depending on the required laser power for absorption. By optimized engineering of the experimental arrangement, we demonstrate how the achievable reduction of pulse-to-pulse energy fluctuation translates into an improvement of the repeatability and resolution in material processing.

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Ablative versus non-ablative laser-induced periodic surface structures

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Laser irradiation of solids can lead to the spontaneous formation of periodic surface structures. This effect is known as laser-induced periodic surface structures (LIPSS). In the last decades, LIPSS have attracted a big fundamental interest, because they gained a broad spectrum of practical perspectives [1]. Therefore, revealing the mechanisms of LIPSS formation and the ability to control surface structuring will allow to develop advanced nanoscale devices.

We have studied the process of LIPSS formation following single-pulse irradiation of pre-existing structures (a step edge or a single groove) on a gold surface for different laser wavelengths. Experimental results are compared with simulations in the framework of a molecular dynamic - two-temperature model (MD-TTM) [2], where laser energy deposition was described by an analytical source term [3], which takes into account excitation of surface plasmon polaritons (SPP) [4] and their interference with the incoming light. Formation of LIPSS is directly observed in ablative and non-ablative laser fluence regimes with differing topographical features, respectively. A powerful MD-TTM tool with a SPP-dependent source term is able to describe LIPSS formation on an atomic scale with a great further potential for controlling surface nanopatterning.

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Modelling sub-micrometer periodic surface structures formation through ultrashort pulsed direct laser interference patterning

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Direct Laser Interference Patterning (DLIP) with ultrashort laser pulses (ULP) represents a precise and fast technique to produce tailored periodic sub-micrometer structures on various materials. In this work, an experimental and theoretical approach is presented to investigate the previously unexplored fundamental mechanisms for the formation of unprecedented laser-induced topographies on stainless steel following proper combinations of DLIP with ULP. The combined spatial and temporal shaping of the pulse increases the level of control over the structure features whilst it brings new insights in the structure formation process. DLIP is aimed to determine the initial conditions of the laser-matter interaction by defining an ablated region while double ULP are used to control the reorganisation of the self-assembled laser induced sub-micrometer sized structures by exploiting the interplay of different absorption and excitation levels coupled with the melt hydrodynamics induced by the first of the double pulses. A multiscale physical model has been developed to correlate the interference period, polarization orientation and number of incident pulses with the induced morphologies. Special emphasis is given to electron excitation, relaxation processes and hydrodynamical effects that are crucial to the production of complex morphologies.

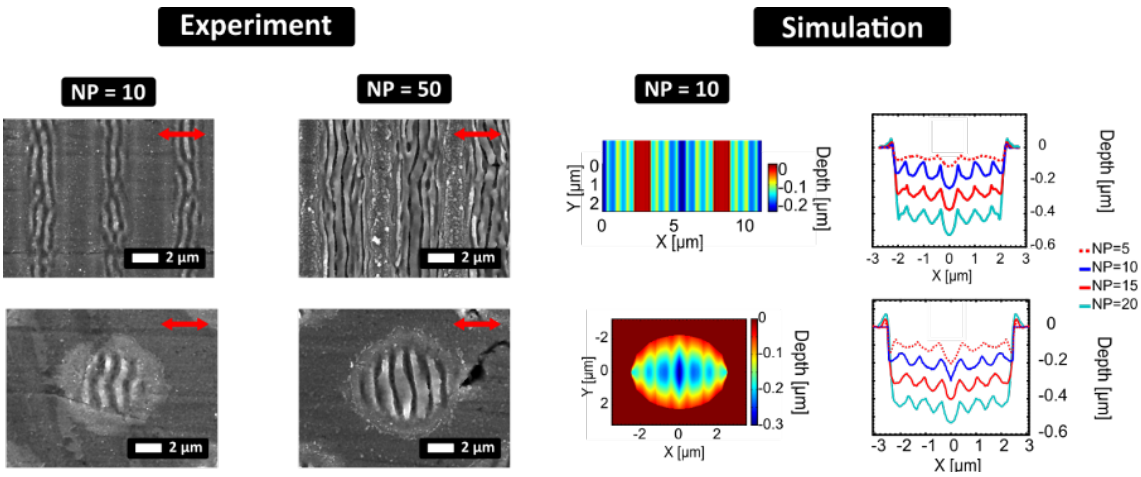

Figure 1: Experimental and theoretical data on stainless steel irradiation by spatially shaped double femtosecond laser pulses. Results are illustrated for two-DLIP (first row) and four DLIP (second row).

Results are expected to derive new knowledge of laser-matter interaction in combined DLIP and ULP conditions and enable enhanced fabrication capabilities of complex hierarchical sub-micrometer sized structures for a variety of applications.

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Cu and Cu-Au electrodes for enzyme-free sensing fabricated by laser-induced selective electroless plating

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It is difficult to overestimate the importance of monitoring various bioanalytes in human blood. Typically, for this purpose are used sensors which are based on electrochemical or optical glucose detection. In the electrochemical approach, this process is typically realized by enzymatic oxidation of glucose. In the case of non-enzymatic detection, the use of enzymes is not required, and oxidation of an analyte (e.g., glucose) is achieved via catalytic ox-red reactions taking place on the surface of the highly developed electrode structure. Here, we show that the method of Selective Surface Activation Induced by Laser (SSAIL) can be used for the fabrication of metallic and bimetallic structures based on copper and gold on the surface of glass and glass-ceramics. It was shown that the fabricated electrodes are suitable for non-enzymatic detection of biologically essential analyte such as glucose.

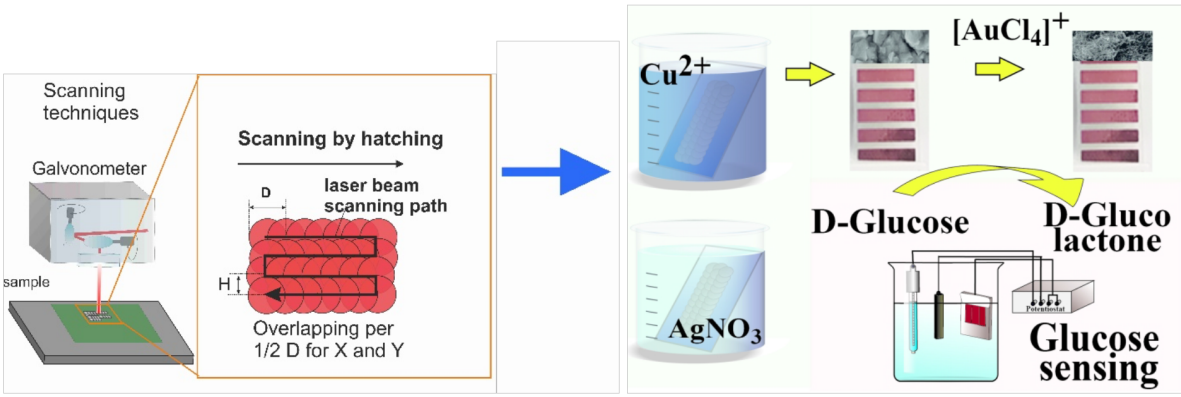

Figure 1: The experimental scheme of laser modification of the dielectric surfaces and their metallization by copper. Step 1: Laser modification of the surface to be metallized with Nd: YVO4 pulse picosecond laser, wavelength 532 nm. Step 2: Activation with silver nitrate solution. Step 3: Electroless copper plating.

The electrocatalytic activity of the fabricated copper and copper-gold electrodes concerning non-enzymatic sensing of glucose was tested using voltammetric methods. It was found that the modification of the copper surface with gold nanoparticles can significantly increase the sensitivity of the electrode towards glucose sensing (3060 vs 911 µA mM^{-1} cm^{-2} for copper on glass-ceramics), improve the detection limit and expand the linear range of detection of this analyte.

I.I.T., E.M.K. and M.S.P. acknowledge Russian Science Foundation (grant 20-79-10075). The authors would like to thank the SPbSU Nanotechnology Interdisciplinary Centre, Centre for Optical and Laser Materials Research and Centre for X-ray Diffraction Studies.

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Internal laser writing inside semiconductors by ultrafast pulses

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Intense infrared lasers can be focused inside semiconductor materials to induce localized modifications arbitrarily in the three-dimensional space. This holds strong potential to integrate multiple functions on a single micro-electronic chip including photonics (waveguide/grating/lens), microfluidic (cooling channel), and mechanics (MEMS), etc. To meet these strong technological perspectives, there are today continuously increasing efforts to improve the precision and stability of 3D laser writing of semiconductor (LWS). Different laser pulse durations ranging from few tens of femtoseconds to nanoseconds have been investigated in this context. Among them, the shortest pulses are of strong interest because they must allow achieving high precision and refractive index writing that are not possible without relying on ultrafast material responses. This is also supported by similar developments made in dielectrics during the last decades. However, previous research shows that ultrashort laser pulses suffer from much stronger nonlinear effects inside semiconductors than in dielectrics. These severely deteriorate the applied laser focusing conditions and cause strong difficulties to exceed permanent modification thresholds.

In this talk, first we give an overview on recent literature regarding ultrafast LWS using pulse duration smaller than 1 ps. A comparison among the literature shows that some inconsistencies persist on the appropriate conditions for LWS [1]. To solve this contradiction, we reveal how the temporal contrast of the pulses should not be ignored as it leads to a laser-technology-dependent conditions for LWS. This is supported by modification experiments showing that femtosecond pulses with contrast as low as 10^{-6} can lead modifications inaccessible with high-contrast femtosecond pulses. To reveal the underlying mechanisms associated with ultrafast LWS, we measure the microplasma induced inside Si and GaAs by ultrafast shadowgraphy and luminescence imaging [2]. The latter reveals in particular a large-scale low-density plasma formed the prefocal region that strongly affecting the energy delivery to the focus.

After confirming the difficulties of ultrafast LWS, we propose solutions to cross the threshold conditions for LWS with high-contrast ultrafast lasers. Our first proposed solution relied on hyper-focused femtosecond pulses (sub-100-fs) to achieve permanent modifications inside silicon [3]. For more practical solutions, we propose new alternatives based on temporal shaping of the ultrafast pulses. First, we investigate the writing conditions with stretched pulses up to the picosecond regime [4]. This allows us to report on the minimal pulse duration for LWS with conventional machining configurations. Another approach is to rely on transient accumulation strategies. To this aim, we generate and apply ultrafast trains of pulses at repetition-rates up to THz [5]. Taken these approaches together, we introduce unique multi-timescale control parameters exploited for improved energy deposition and for demonstrations of reliable 3D laser writing deep inside silicon chips that would not be possible otherwise.

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Atomistic simulations of the generation of alloy nanoparticles by short pulse laser ablation in liquid

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The ability of short pulse laser ablation in liquids (PLAL) to produce colloidal solutions of chemically clean nanoparticles has been employed in a broad range of practical applications. Large-scale atomistic simulations have yielded important insights into the fundamental mechanisms of PLAL [1-4] and provided a plausible explanation of the origin of the experimentally observed broad or bimodal nanoparticle size distributions. In the computational effort reported in this presentation, we extend the atomistic simulations to investigation of the nanoparticle formation mechanisms in PLAL of Ag/Cu and Cu/Ag bilayer thin films [5], as well as FeNi alloy targets. The computational predictions are related to the results of experimental studies performed at the University of Duisburg-Essen and Kiel University, Germany.

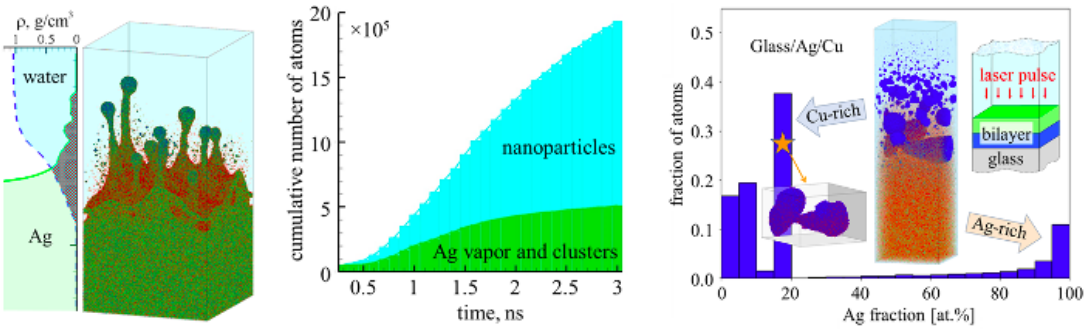

Figure 1: Example illustrations of the results of atomistic simulations of laser ablation in liquids.

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Single-shot formation of hollow microneedles in silicon by doughnut-shaped laser pulses

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We report on the formation of tubular-like structures and hollow-core microneedles on the surface of monocrystalline silicon using ultrashort laser pulses. Highly deterministic surface processing is ensured by single-shot ablative modification of the sample surface above the laser-induced damage threshold using radially polarized doughnut-shaped laser pulses with a duration of 35 fs (Astrella from Coherent, 800 nm wavelength). At laser fluences slightly above the ablation threshold, well reproducible tubular structures are formed whose height increases with fluence culminating with closing the structure on the top (fig. 1). Upon multi-pulsed irradiation, the height of the needle structures can be increased as compared to those produced by single pulses but, at certain number of pulses, melting and ablation cause the entire structure to collapse. The mechanisms responsible for creating these surface structures are discussed based on thermodynamics, hydrodynamics, and material stress theory. The results of our systematic study of the formation of hollow structures and their top closure can open new opportunities for applications in silicon photonics, optical field enhancement on microstructures and silicon-based photovoltaic devices.

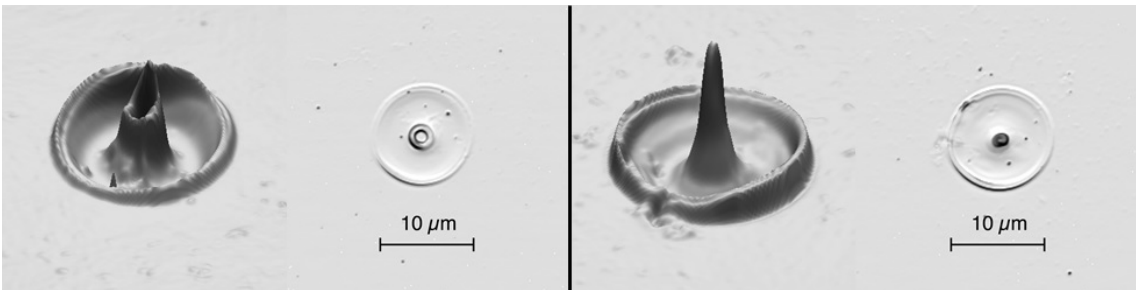

Figure 1: Confocal microscopy images of a tubular-like structure (left, 3D and top images) and hollow-core microneedle (right, 3D and top images) on the surface of monocrystalline silicon created by single doughnut-shaped femtosecond laser pulses.

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Section 4: Femtosecond non-linear optics, filamentation, high field THz generation

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(Zuev Institute of Atmospheric Optics, Russia)

Scope

Self-compression and self-focusing

Terahertz science

Filamentation in various media

Femtosecond nonlinear phenomena

Evolution of a long lived laser guided discharge

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We present an experimental investigation on electric discharges of 80-cm length guided with laser filament. The discharge is created with an electrical circuit composed of two parts. The first one is a Tesla coil generator with maximal voltage amplitude about 300 kV [1]. It generates a HV breakdown into a laser filament prepared channel, followed by a short HF discharge. In this way a plasma channel with high conductivity is created. The second part of the circuit serves for the discharge prolongation. It is composed of a ground coil **L** with inductance $L = 47.8$ mH and a capacitor **C** of different values between ~ 350 nF and ~ 720 nF charged up to $U_0 = 20$ kV. This relatively low voltage value is enough to maintain the guided discharge created in the first stage. By changing the capacitance of the second circuit we are able to change the current discharge value in the range 30–43 A and its duration between 1.75 ms and 3.5 ms. This allowed us to investigate the evolution of the diameter and brightness of the discharge channel as a function of time using a high speed camera.

The FWHM diameter of the plasma channel evolves in a stepwise manner. The first step diameter is less than 1.5 mm and the second step is 3–5 mm, depending of the current. These two levels correspond to the first 2.1–3.5 milliseconds of the discharge, when the current passes through the channel.

We then observe a second phase in the channel evolution after the end of the injected current. This current-free plasma channel keeps a straight cylindrical geometry up to about 10 ms for the 720 nF circuit. After 10 ms we start to note some degradation of the guiding structure and the plasma fluorescence becomes very low.

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High order Brunel harmonics and supercontinuum formed by a weak optical pump in presence of a strong terahertz field

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Brunel harmonics appear in the optical response of an atom in process of laser-induced ionization, when the electron leaves the atom and is accelerated in the strong optical field [1-3]. In contrast to recollision-based harmonics, the Brunel mechanism does not require the electron returning to the core.

Here we show that in the presence of a strong terahertz (THz) field, which is just below the boundary of an effective tunnel ionization, even a weak driving field at the optical frequencies allow for generating Brunel harmonics especially effectively. Importantly, the strong ionizing THz pump also suppresses recollisions, making the Brunel mechanism dominant in a wide spectral range. High-order Brunel harmonics may form a coherent carrier-envelope-phase insensitive supercontinuum, compressible into an isolated pulse with the duration down to 100 attoseconds.

The main idea is presented in Fig. 1a: nonlinear response E_r is generated by electron tunneling from an atom in a strong THz field E_{THz} superimposed with a weak optical pump E_{opt}. In Fig. 1b, the electron wavefunction according to 1D Schrödinger equation is presented for an exemplary case, supporting that the strong THz field suppresses the recollision harmonics.

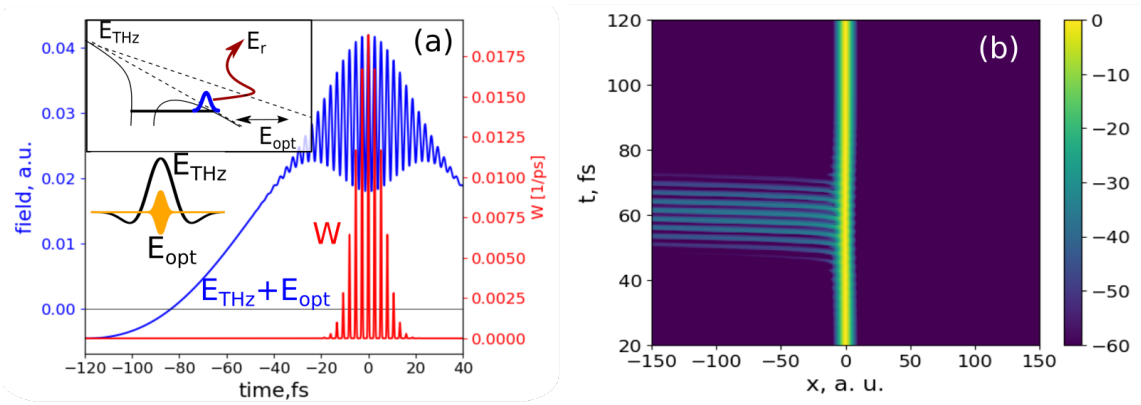

Figure 1: (a) Nonlinear response of an atom to a weak optical probe pulse in presence of a strong THz field – a simple-man picture. (b) Electron wavefunction (in logarithmic scale) resulting from a Schrödinger equation simulation and demonstrating suppression of recollisions in the scheme.

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Ultrafast nonlinear optical studies of central cavity varied novel Phthalocyanines

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In this present investigation, we present data from extensive nonlinear optical (NLO) studies of two novel central cavity varied tri-methoxy-phenoxy-phthalocyanine (TMPC) molecules with femtosecond laser pulses. We have utilized $\sim$ 50 fs amplifier pulses at 1 kHz repetition rate and 800 nm as excitation source. We have extracted the NLO coefficients by probing these compounds using the Z-scan experimental technique using these pulses. The exotic molecules have clearly revealed a saturation absorption combined with reverse saturable absorption nature, exhibiting significant 'M-shaped' curves in open aperture and a clear defocusing Kerr-lens effect in closed aperture configuration. An interesting intensity dependent NLO study has also been carried out, unveiling further exotic nature. These compounds were characterized utilizing the Matrix assisted laser desorption ionization (MALDI) technique and their structures are designed as follows

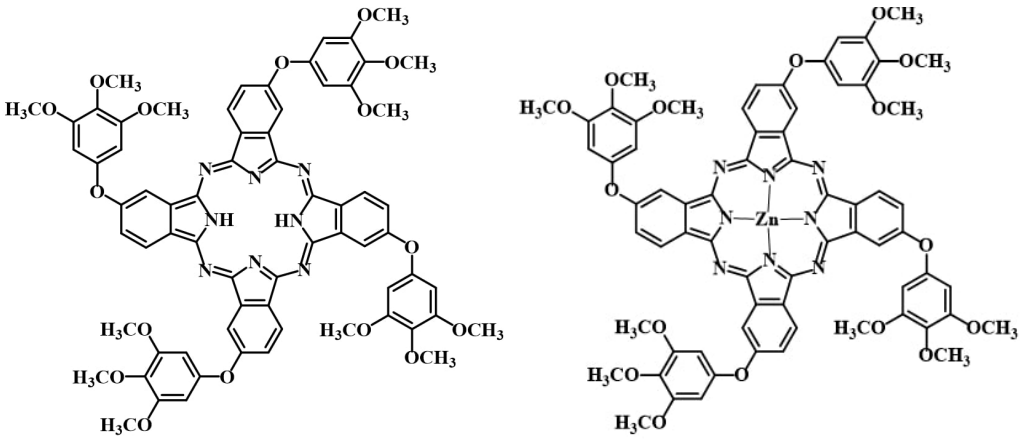

Figure 1: Molecular Structure of TMPC (left) & Zn-TMPC (right) investigated in the present study.

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The second optical harmonic generation at a wavelength of 620 nm in an antiferromagnetic NiO, induced by broadband and narrowband THz pulses

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Nonlinear optical measurements are very sensitive to changes in electrical and magnetic symmetry, crystallographic orientation, and polarization caused by external fields. Therefore, second harmonic generation (SHG) is one of the effective methods for studying the properties of various crystals and thin films. In this report, we present the results of an experimental study of the generation of the second optical harmonic at a wavelength of 620 nm in a centrosymmetric antiferromagnet NiO, induced by broadband and narrowband THz pulses. SHG was investigated using the "pump-probe" scheme in co-propagation geometry, when terahertz pump and probe pulses with a wavelength of 1240 nm propagate in the same direction.

The experiments were performed using the unique scientific facility "Terawatt Femtosecond Laser Complex" in the "Femtosecond Laser Complex" Center of the Joint Institute for High Temperatures of the Russian Academy of Sciences.

The reported study was funded by RFBR, project number 20-08-00627.

THz plasma source angular distribution modification by use of pump beam regularization

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Laser plasma THz sources based on two-color femtosecond pulse focusing attract attention due to their ultrabroad spectrum 0.1–200 THz [1], relatively high peak fields [2], and the possibility of forming a source in front of a studied object avoiding losses due to propagation in air. However, the emission pattern from such a source is complex [3–5]. Therefore, it is important to clarify the optimized condition of THz beam generation and to develop ways to control the spatial distribution of THz emission by the two-color scheme. Here, we experimentally study THz emission pattern from complex laser plasma sources formed by several plasma channels.

The possibility of controlling THz emission spatial distribution for composite laser-plasma source has been demonstrated using amplitude mask for laser beam and introducing delay between beamlets. The interaction of two non-delayed beamlets leads to formation of a complex superfilament-like structure, which is characterized by a bright component of THz emission propagating along the axis. The introduction of delay between two beamlets shifts the maximum in angular and frequency-angular distributions. However, it should be noted that the spatial modulation decreases the total energy of THz emission, as the highest THz yield is observed for the single filament. The ring component, corresponding to THz radiation of the original filaments, is also present. These results can be useful for high power broadband laser-plasma THz sources and for control of the THz pattern.

The study is supported by Scholarship of Russian Federation President (SP-500.2019.4).

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Thriving for highest signal to noise in active sensors: free space versus fibers

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This work refers to sensors based on beating two correlated frequency combs issued from the same Optical Parametric Oscillator (OPO) cavity. Because these combs are correlated, beating them together on a detector leads to a low frequency signal of very narrow bandwidth (at least 6 orders of magnitude smaller than the bandwidth of a mode of either comb). With the free space OPO of Fig. 1, a beat note mean square deviation (MSQ) of $\sqrt{\langle \Delta\nu^2 \rangle} = 0.06$ Hz was measured. For the 20 ns round-trip time cavity, this MSQ corresponds to a phase uncertainty of $\sqrt{\langle \Delta\phi^2 \rangle} = 0.06 \times 2\pi\,(20 \cdot 10^{-9}) = 0.8 \cdot 10^{-8}$, which is close to the quantum limit. It corresponds also to a sub-fm sensitivity in optical path. The possibilities of a sensor enhancement that is not subject to the same excess noise buildup that is observed in Exceptional Point based enhancements [1] is presented.

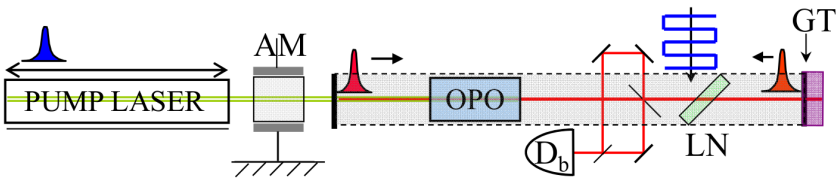

Figure 1: An OPO cavity is pumped at twice the repetition rate of its cavity. The amplitude of alternate pump pulses is controlled by an electro-optic modulator *AM*. *LN*: phase modulator driven at the round-trip frequency of the OPO, simulating the signal to be detected. D_b: beat note detector. *GT* Resonant dispersion element (Gires-Tournois) for signal enhancement.

It is obvious that fiber implementation promises a considerably reduction in energy consumption volume and weight. Fiber laser gyros have been extensively studied by Krylov *et al* [2]. We investigate a ring OPO and a linear OPO. Figure 2 shows the beat note recording in time Fig. 2(a) and frequency Fig. 2(b). In comparison to the free space OPO, the beat note bandwidth is considerably broader. The origin of additional noise and possible remedies will be discussed.

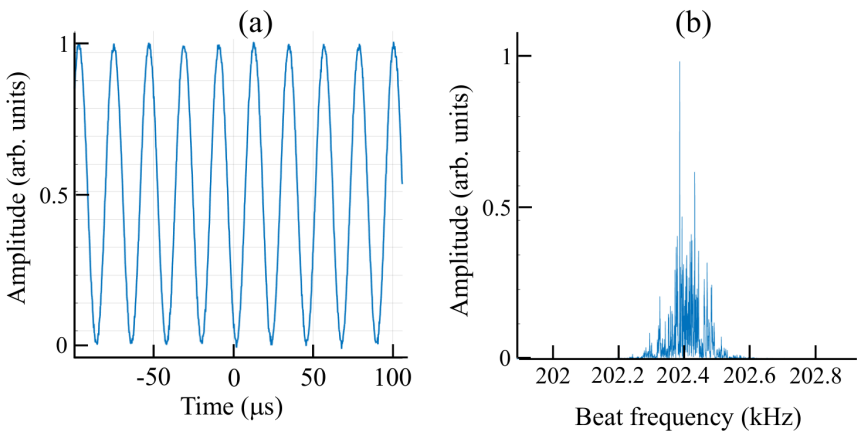

Figure 2: (a) Recording of beat note of ring OPO in time. (b) Fourier transform of the beat note time trace.

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Optimized femtosecond laser-driven K_α hard x-ray generation from Cu target placed in gas under overcoming the impact of clamping intensity regime: towards diffraction experiments and backlight imaging.

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We present an experimental approach for time-resolved X-ray experiments based on a femtosecond laser-driven microplasma K_α X-ray source. The employed design provides flexibility to adapt the setup to such requirements as sample environment and X-ray optics. The developed scheme has been optimized toward gas environment. This makes it possible to perform dynamic measurements of strain waves triggered by a femtosecond laser, diffraction measurements of the non-stationary structural dynamics used crystalline materials, and can also be used as a micron source of X-ray radiation the backlight imaging for materials science problems.

We discuss peculiarities of tight focusing conditions to produce intense hard X-ray source. It will be shown that intensity clamping due to plasma generation and nonlinear losses does not essentially limit the femtosecond laser matter coupling, thereby giving access to a new propagation regime featured by an optimized laser energy deposition in the target and exceeding pulse intensity 10^{14} W/cm^2 at the focus.

A phenomenological analysis of the features of X-ray generation in the optimal laser-matter interaction regime when the target is placed in the air accompanying the conditions of local helium jet injection into the working zone by a tightly focused femtosecond laser beam, has been carried out.

In this study, we present experimental investigation on the interaction of repetition rate (10 Hz) TiSa femtosecond laser focused on the target by an Off-Axis Parabolic (OAP) mirror with 50 mm focal length ($NA = 0.07$). The dependence of X-ray yield on the femtosecond pulse duration and laser energy is investigated. The experiments performing with K_α-radiation (8 keV) from a Cu foil target, whose surface is covered by a helium curtain using directed helium jet. It is found that the X-ray yield reaches the maximum of $2.5{\cdot}10^7$ phot/pulse in 2π sr (or $2.5 \cdot 10^8$ phot/sec) in 2π sr at laser energy of 6 mJ and at pulse duration of 300 fs. This value differs from the X-ray output obtained under vacuum conditions at comparable femtosecond laser pulse energy less than an order of magnitude. In the regime of the transform-limited pulse X-ray yield is minimal (30 fs) and is about 10 times lower than in the maximum (see Fig. 1a). This is because we assume that the intensities in focus for pulses with different durations due to clamping are almost the same, but the discrepancy in pulse durations results in the same disparity in energy density.

In the experiments carried out, an accompanying X-ray production second harmonic of laser radiation was observed. The spectrum of which is blue shifted. The SHG signal is maximal at 300 fs and disappears in the generation regimes of long pulse duration (2 ps). In the transform-limited mode (pulse duration 30 fs) only the supercontinuum emerging in He layer was observed.

Using the characterized X-ray source, the diffraction experiments with Si target have been performed. We apply the scheme to highlight a high-contrast copper line in the broadband X-ray spectrum using crystalline silicon (111). The diffracted X-ray at the sample situated at

Bragg angle is recorded with high contrast respect to background (see Fig. 1b). The probing X-ray radiation emitted over a range of 2π sr was collimated using slits of 1×20 mm^2.

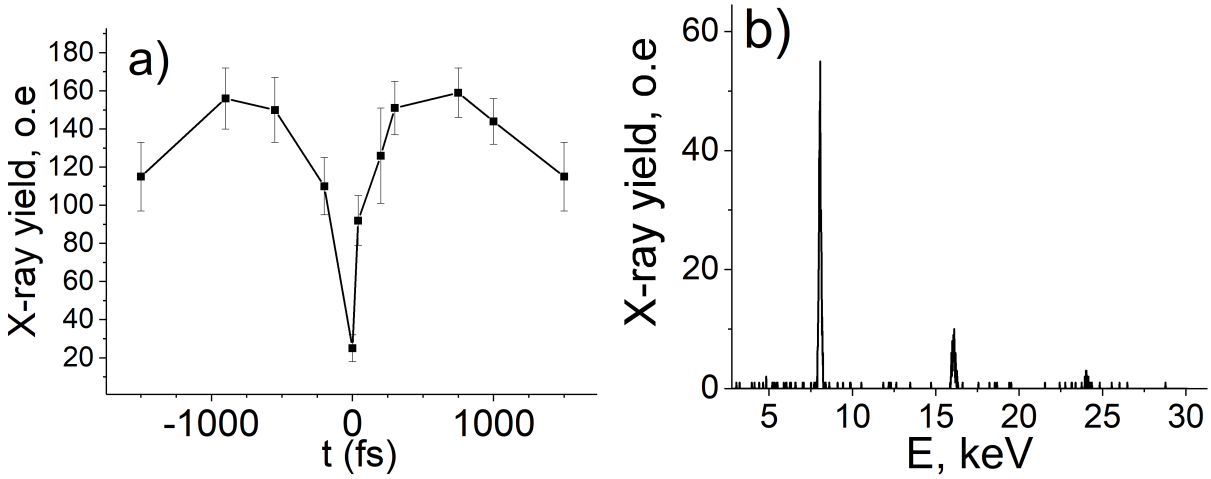

Figure 1: Dependence of the X-ray yield on the pulse duration under focusing with $NA = 0.07$ at laser pulse energy of 6 mJ. Point 0 on the time scale is corresponding to the transform-limited pulse of 30 fs (a). X-ray diffractive line from (111) Si measured with spectrometer Amptek (b).

The work was supported by the Russian Foundation for Basic Research (RFBR 18-29-20074, 18-02-40018, 18-02-40032, 19-29-12037).

Femtosecond filamentation of structured light: Perspectives for long-range pulse propagation

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The unique properties of high-power laser pulses, leading to nonlinear optical filamentation phenomena, higher harmonic generation, supercontinuum luminescence, and the formation of elongated plasma channels, make them attractive not only for laser technology problems, but also for solving atmospheric optics problems associated with directional laser energy transmission [1]. One of the main problems of femtosecond atmospheric optics is the control of the multiple filamentation region of high-power radiation along the propagation path from the point of view of the formation of filaments and plasma channels at a given distance from the laser source (hundreds and thousands of meters). As demonstrated, the manipulating of laser filamentation by changing its wavefront (spatial focusing) is the most effective way to control the power and spatial parameters of pulse energy concentration region during nonlinear propagation.

At the same time, on the long-range atmospheric links for multi-gigawatt and terawatt laser pulses, the use of traditional focusing optics to control the filamentation becomes challenging because of very random nature of this nonlinear phenomenon. The stochastic nature of multiple filamentation introduces strong variability in the spatial position, number and transverse dimensions of plasma-free light channels organized at the post-filamentation stage. Undesirable stochastic multiple filamentation of a high-power laser pulse can be prevented by making this process regular. This is usually achieved by a special modification of the amplitude-phase profile of high-power radiation before sending it into a nonlinear medium by using various aperture masks, aberration, segmented focusing, or deformable flexible mirrors, adaptive optics. Another method for high-power pulse propagation control involves the use of specially profiled radiation, i.e. laser beams with non-unimodal (non-Gaussian) transverse intensity distribution. The practical interest in the spatially structured radiation is associated with the specific features of the linear diffraction of such beams, which in some cases leads to a delay in the nonlinear focus formation.

In this contribution we report the results of our recent theoretical and experimental studies on the multiple filamentation in air of high-power ultrashort laser radiation with various structured intensity profiles. The structured beam profiles were obtained by different methods through amplitude and/or phase pulse profiling and resembled a "dressed beam", a π-shifted segmented beam, a "corona-beam" composed of several annularly distributed sub-beams. We studied the spatio-angular dynamics of structured femtosecond pulses along the optical path by varying the number and peak power of the beamlets. The evident advances in the multiple-filamentation region manipulating of structured beams are demonstrated. Particularly, by adjusting the number and aperture of the constituting sub-beams it makes possible to significantly delay the filamentation onset distance and increase the filamentation length in air. In addition, at the post-filamentation stage of the femtosecond pulse propagation under certain conditions the structured beams exhibit significantly lower angular divergence of its most intense part compared to the beams with regular unimodal intensity profiles that provides enhancing of laser power delivered to the receiver on atmospheric links.

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Conversion of Ti:Sa laser radiation due to spectral broadening in post-filament channel and intrapulse difference frequency generation

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Nowadays, sources of coherent ultrashort pulses in the mid-infrared (MIR) range are vital for femtochemistry [1], multidimensional spectroscopy [2] and attosecond pulses generation [3].

One of the most convenient ways to obtain coherent radiation in the MIR range is intrapulse difference frequency generation (IDFG). This technique does not demand the use and alignment of two beams, but a wide (tens of nanometers) spectrum is essential. We suggest to apply spectrum broadening in the post-filament channel [4] for Ti:Sa output. Broadened spectrum has humps with a distance that can be varied by pulse duration, pulse energy and numerical aperture (NA) adjusting.

For IDFG we use LiGaS2 since it has wide spectral acceptance at $\sim$ 11 µm wavelength and large bandgap energy. Broadening required for IDFG at 11 µm (fig. 1 (left)) is observed for pump at the 744 nm with initial energy of 1.8 mJ, duration of 100 fs and $NA = 0.001$.

Spectrum of MIR signal (fig. 1 (right)) is measured by homemade spectrometer. The energy efficiency is estimated as 10^{-4} and $5 \cdot 10^{-4}$ with and without chirp compensation respectively. The spectrum width corresponds to the transform limited pulse duration of 47 fs.

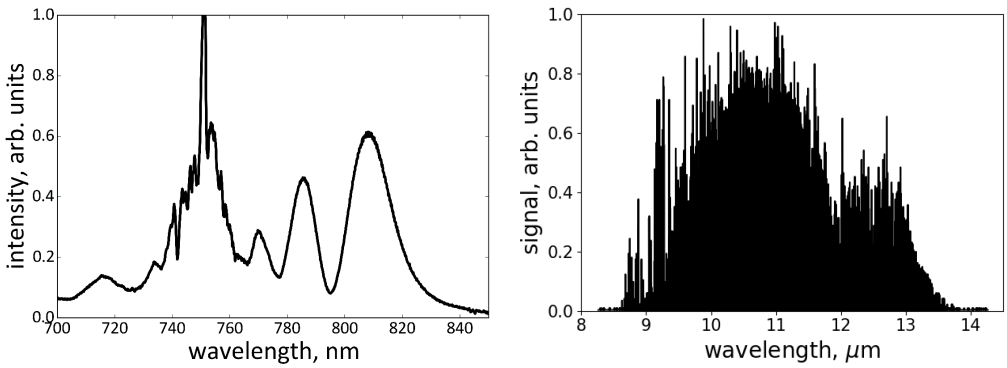

Figure 1: (left) Spectrum of the post-filament channel; (right) MIR signal.

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Liquid jet based broadband terahertz radiation source

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The development of systems for the generation and detection of broadband terahertz (THz) radiation is of great interest both from an applied and a fundamental point of view. The approach based on the of laser pulse filamentation in various media is gaining high popularity. The search for the optimal media and conditions for pulsed pump radiation is an important issue.

In this work, the system for generating a liquid jet is based on the nozzle previously used for dye lasers [1] and then applied for linear time-resolved THz spectroscopy systems (operating at low power) to study the liquid properties [2], as well as for plasma-based x-ray generation [3]. In work [4] the detailed description of the liquid jet formation system (included in the THz radiation generator) was present. Highly efficient liquid-based THz sources are also studied extensively in work [5].

This work provides a complete description of a liquid jet terahertz radiation generator using a plane-parallel jet, consisting of a liquid jet formation system and an optical part. The applications of this generator for studying the optical-to-terahertz conversion efficiency for various liquids as well as THz energy temperature dependence are demonstrated. This system allows to achieve the optical-to-THz conversion efficiency up to 0.1% in the case of α-pinene double-pulse excitation.

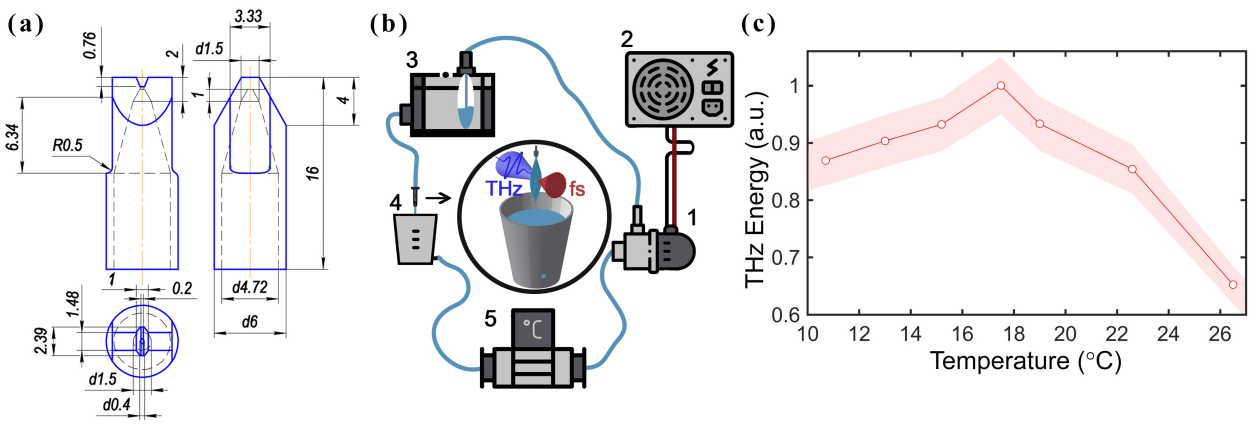

Figure 1: (a) The schematic drawing of a nozzle for producing the liquid jet (the dimensions are given for 100 µm water jet thickness). (b) The system of jet formation, which is built into the optical setup: 1 — pump, 2 — power supply, 3 — damping capacity, 4 — nozzle with liquid collecting capacity, 5 — chiller. (c) The temperature dependence of THz emission generated in case of single-pulse excitation of water jet.

The study is funded by RSF grant № 19-12-00097.

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Light bullets. Spatio-temporal and energetic characteristics.

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During the filamentation of a femtosecond laser radiation under anomalous group velocity dispersion, a light bullet occurs – an extremely compressed wave packet with a high light field localization both in space and in time. The report briefly shows the state of the art on the investigation of light bullets formed in condensed media and air. The existing estimations of light bullet's parameters are discussed [1,2].

Based on the analysis of the spatio-temporal distribution of the electric field calculated in the approximation of unidirectional femtosecond radiation propagation, one defined the localization region of a high light field. The duration, radius and energy of this region are absolute parameters of the light bullet. It is found that the duration of the light bullet in LiF is independent on the wavelength and equals to 1.8 optical cycles. Its radius equals to three wavelengths in the region of a strong anomalous group velocity dispersion and decreases to one wavelength in the vicinity of zero dispersion region. The energy in the light bullet aperture increases more than 10 times during the light bullet formation and reaches 1.5 µJ.

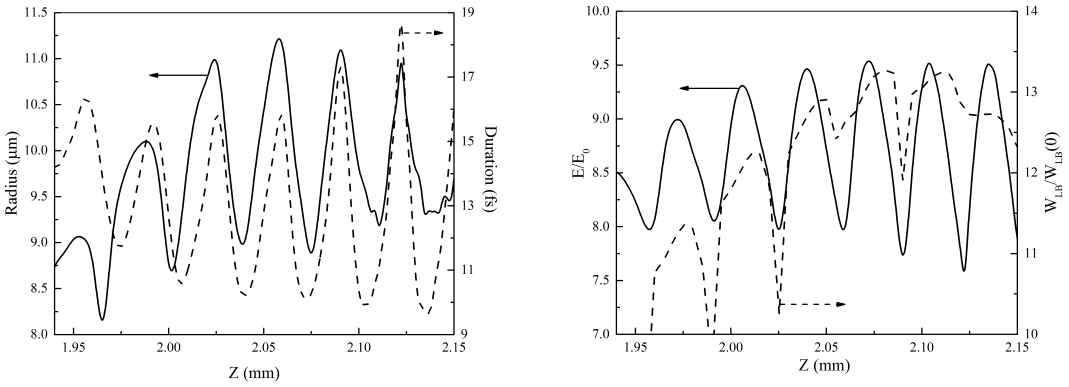

Figure 1: Oscillation with propagation of parameters of light bullet at $\lambda_0 = 3100$ nm.

The parameters of a formed light bullet periodically change with the propagation as a consequence of the shift of an electric field's absolute phase [3]. The cophased compression of the light bullet in space and time is accompanied by an increase in a peak electric field strength. The growth of the energy in the light bullet occurs simultaneously with its cophase spatio-temporal broadening (Fig. 1).

Experimentally one investigated the oscillations of light bullet's parameters by the use of laser coloration technique. This technique allows to register in the single-pulse regime changes in the amplitude and distribution of the electric field in light bullet during its formation and propagation in the filament. Introduced absolute parameters of the light bullet are in line with characteristics, reconstructed from measurements of induced by it structures of color centers.

The study was supported by a grant of the Russian Science Foundation (Project No.18-12-00422)

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Model of micromodification induced by multi-pulse filamentation in LiF

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Lithium fluoride (LiF) is a unique material for studying the process of femtosecond laser filamentation due to the formation of stable fluorescent color centers (CCs), such as F_2 and F_3^+ CCs, in irradiated areas [1-3]. Using LiF, it is possible to experimentally study fluorescent tracks of both single-pulse and multi-pulse filamentation. One of the experimentally observed facts is the elongation of the tracks in LiF during multi-pulse filamentation [4,5]. This work is devoted to the study of the nature of this phenomenon and the development of a mathematical model that reproduces the observed modification of LiF.

The developed model takes into account two channels of CCs formation: excitonic and electron-hole channels. The relative contributions of these channels are determined by the accumulated CCs density. The model describes a permanent modification of the refractive index in irradiated areas of the material, where CCs are present. Each subsequent laser pulse propagates in a medium modified by all previous pulses.

The model was used to simulate a waveguide structure formation during multi-pulse filamentation at a wavelength of 3 µm, located in the spectral region of anomalous group velocity dispersion. Results of the simulation qualitatively reproduce the experimentally recorded elongation of the fluorescent tracks of filamentation.

This work was supported by the Russian Science Foundation, project №18-12-00422, and the Basic Research Plan of the Russian Academy of Sciences for the period up to 2025, project № 0243-2021-0004.

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Lasing of molecular nitrogen ions in laser plasma

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Lasing on first negative system of molecular nitrogen ion (transition $B^2\Sigma_u^+$–$X^2\Sigma_g^+$) emitting at wavelengths of 391,4 and 427,8 nm in forward direction was obtained in 2011 [1] by pumping an intense ultrashort laser pulse in ambient air. Cavity-free generation of coherent radiation holds great potential for remote sensing applications. One of the most important parameters of lasing is pulse duration. In first studies it is believed that lasing pulse duration should be transform-limited ($\sim$ 1 ps) [2]. Soon afterwards, cross correlation technique and direct measurement by streak camera with high precision were performed to determine the lasing pulse duration. It was found that lasing pulse duration could be more than that of transform-limited (2 — 7.8 ps) [3].

Typically, studies, devoted to the determination of lasing pulse duration from molecular nitrogen ions, were carried out under the same experimental conditions, namely the pump duration, pump energy, and lens focal length. The conditions of air lasing at different laser plasma length, gain coefficient value, etc. were not investigated. Almost all researchers speak about the amplification of femtosecond seed pulse or about the amplification triggering by seed pulse. However, there is no explanation of the fact, that the lasing duration has the picosecond duration.

This work is devoted to the investigation of lasing pulse duration behavior in air and nitrogen chamber (λ =428 and 391 nm) at different focusing conditions of the laser emission. Pumping was carried out at the wavelength of 950 nm and had pulse duration of 50–60 fs. Analysis of active medium parameters under these conditions was performed. The mechanism, responsible for the formation of picosecond lasing pulse duration, was suggested.

The reported study was funded by RFBR according to the research projects № 20-08-00060 and 19-48-700016.

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Femtosecond laser induced photoelectron emission from nano graphite flakes photocathodes

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Generation of ultrafast electron bunches is of great significance in a wide variety of applications in the fields of vacuum electronics and generation of THz pulses. Laser induced photoelectron emission from different materials is one of the main picoseconds and subpicoseconds electron pulses generation methods [1,2]. There are plenty of high-performance photocathodes for the visible and infrared ranges, but their sensitivity to the quality of vacuum is high, that's why their usage in a wide range of physics experiments is difficult. Therefore, atmosphere tolerant photoelectric materials search is vital task nowadays. In order to generate an ultrashort electron pulse, we use a simple compact structure with an accelerating field (∼10 kV/cm) between a mesh anode and a photocathode in a vacuum chamber. Laser radiation passing through the mesh anode hits the photocathode and initiate photoelectrons. Photocathode material selection is based on the possibility to initiate the photoelectric effect for these materials. A plain metallic photocathode exhibits photoelectric properties with a fairly low efficiency. NGF (Nano Graphite Flakes) demonstrates rather high thermal conductivity that exceeds, for example, copper five times. This fact allows us to expect a higher yield of electrons. The most advantage of these photocathodes is its capability to operate with these photocathodes at ambient.

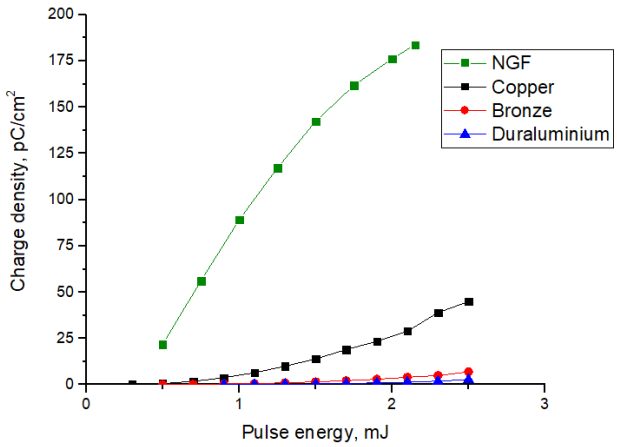

Figure 1: Measurements of charge density.

We studied the charge emission during photoelectron discharge of the photodiode on the laser pulse energy for metal and NGF samples (Fig.1). At any pulse energy the value of the charge density for a copper photocathode is higher than for any studied metal ones. Our experiment results demonstrate 4 times profit for charge density for NGF-photocathodes comparison with plain metal ones.

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Multiple filaments and superfilaments of femtosecond terawatt laser pulses over an extended atmospheric path

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We present the results obtained using a new experimental set up for long multiple and superfilaments studies. The filaments were formed on atmospheric path with the length about 50 m and the set up permits us to work with laser pulses with energy up to 50 mJ. We performed measurements at many distances along the path in case of stochastic, regularized with amplitude and phase masks filaments and observed a stable filament array formation in case of amplitude masks.

We used Ti:Sa laser system (805 nm, 10 Hz, beam diameter (FWHM) 14 mm, energy up to 50 mJ, pulse duration 55 fs). We created a vacuum path to deliver fs radiation to a direct extended path (corridor) and a mobile diagnostic utility, that implements broadband acoustic diagnostics (bandwidth – 6 MHz), and spatial mode (intensity distribution observed crossing the beam with a paper) diagnostics. The imaging angular spectrometer and THz diagnostics can also be placed on this stage. We considered different focusing condition and also investigated the filament formation without external focusing.

In particular, we studied multiple filaments regularized with different amplitude masks. The filament dynamics is presented in Fig. 1. We observed a stable filament array, that has been preserved over almost 10 m. The filaments in array are arose almost simultaneously. The volume energy density in this regime deduced from the acoustic signals amounts to 0.0025 J/cm^3. In case of stochastic filamentation the first filament appeared 10 m from the beginning of the path, filaments length was 10-15 m. The filaments formed one after another.

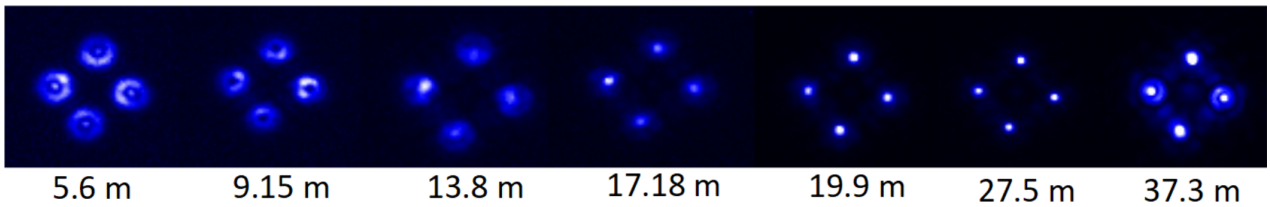

Figure 1: Mode images of the beam after the amplitude mask with four holes.

The reported study was funded by RFBF according to the research projects #18-52-16020, #18-02-00954, #20-31-70001 (E. V. Mitina), funded by RSF project #21-12-00109, BASIS Foundation for the Advancement of Theoretical Physics and Mathematics (19-2-6-261-1, E. V. Mitina).

Strong field THz generation in a non-phasematched, large area $LiNbO_3$

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Single-cycle terahertz (THz) pulses with a high-intensity electric field can be obtained by the conversion of femtosecond laser radiation. For pump-probe experiments in strong fields and with a 30 fs resolution, a simple method is required to obtain high-power, short THz pulses from 0.8 µm multiTW laser systems. We demonstrated that the well-known lithium niobate crystal (LN) has advantages in this situation, despite strong THz absorption and the absence of phase matching [1].

The advantages of LN are high damage threshold together with high nonlinearity [2], as well as the availability of crystals of large area and small thickness. The high nonlinearity makes promising the simplest configuration of THz generation – normal incidence onto a thin plate. High thresholds of damage and THz yield saturation allows to use high intensities of laser pumping, which increases the efficiency in this quadratic generation process. The large available area makes it possible to obtain a high THz pulse energy with a sufficient power of the laser system. Non-phasematched generation in a thin layer provides significantly broadband and short THz pulses in comparison with other methods [3]. In this work we optimized intensity and duration of the laser pulse, thickness and area of the crystal to obtain a strong THz field in the LN. We also investigated THz and visible spectrum modifications. As a result with 250 mJ of laser energy, we obtained a 10 µJ, single-cycle THz pulse with a broad, smooth spectrum, containing 40% of the energy at frequencies above 3 THz. That is twice more energetic THz pulse than obtained in a rare gas plasma pumped with the same laser system, but in a two-color scheme [4].

This work was partially supported by RFBR grants 18-52-16024, and 18-02-40032.

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1D and 3D Maxwell-Bloch modelling of N_2 and N_2^+ lasing in plasma filaments

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Backwards amplification of UV radiation in atmospheric filaments has a huge potential as a source of intense, directional and coherent probe pulses for remote sensing applications. The swept-gain nature of these amplifiers results in a weak backwards signal that requires a complex optimization process in order to be used in applications of interest.

In this presentation we will show 1D and 3D modelling of the amplification of UV radiation in Nitrogen filaments. The amplification mechanism in both neutral Nitrogen molecules and ionized Nitrogen molecules will be studied. We will show the role that plasma dynamics, through electron collisions, plays in shaping the amplified beam spectral and spatio-temporal profiles

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Broadband and narrowband laser-based terahertz source and its application for excitation of ferroelectric

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The ideal laser source for nonlinear terahertz spectroscopy offers large versatility delivering both ultra-intense broadband single-cycle pulses and user-selectable multi-cycle pulses at narrow linewidths. Here we show a highly versatile terahertz laser platform providing single-cycle transients with tens of MV/cm peak field as well as spectrally narrow pulses, tunable in bandwidth and central frequency.

The processes of dynamic polarization switching induced by the strong electric field of a nearly single-cycle terahertz pulse are studied in a classical ferroelectric silicon-doped lead germanate single crystal, $Pb_5(Ge_{0.74}Si_{0.26})_3O_{11}$, in THz pump – second harmonic generation probe geometry depending on the pulse electric field strength.

The experiments were performed using the unique scientific facility “Terawatt Femtosecond Laser Complex” in the “Femtosecond Laser Complex” Center of the Joint Institute for High Temperatures of the Russian Academy of Sciences. The reported study was funded by RFBR and ROSATOM according to the research project № 20-21-00043.

Frequency-angular structure evolution of postfilament channel on 100 m atmospheric trace

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We observe the formation of relatively stable postfilamentation light structure during the propagation of 100 fs, 5 mJ, 744 nm laser pulse in ambient air both experimentally and in numerical simulations. In order to characterize such postfilament channel we measured frequency-angular spectra, autocorrelation functions and beam profiles at the distances up to 95 m. It is shown that in the range of distances from 65 m to 95 m postfilamentation channel diameter and frequency-angular characteristics (see fig. 1) remain almost constant and insensitive to initial energy variations.

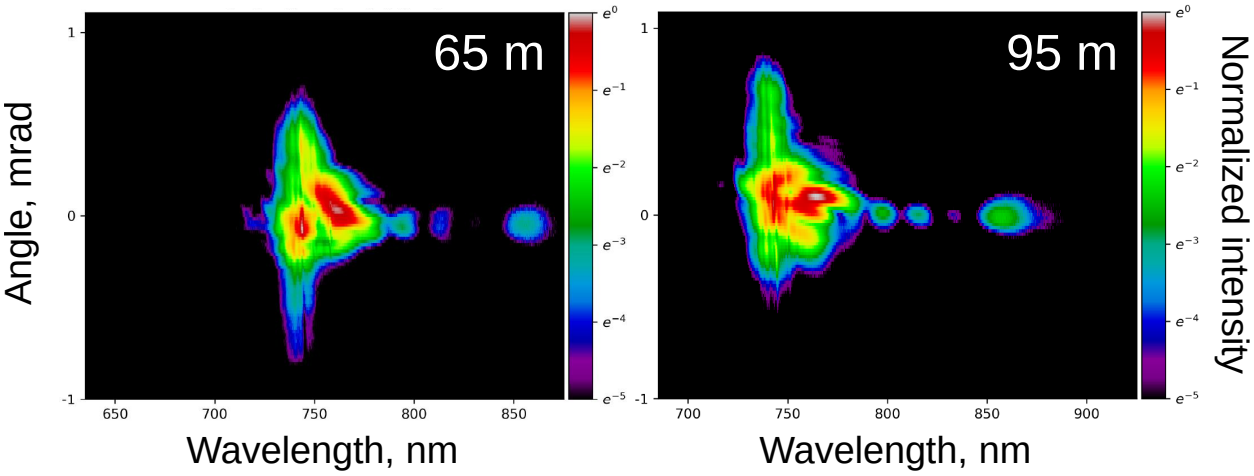

Figure 1: Experimentally obtained frequency-angular spectra taken at the distances of 65 m (left) and 95 m (right) from the laser system.

This research was funded by Russian Science Foundation, grant № 21-12-00109.

Controlled resonant third harmonic generation in clusters with intense femtosecond laser

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Third harmonic generation (THG) in a cluster jet occurs due to the optical nonlinearity of nanoplasma induced by high-intensity ($I > 10^{15}$ W/cm^2) femtosecond laser radiation. It is known, when the plasma frequency of the ionized cluster ω_p becomes equal to the tripled frequency of the incident laser radiation 3ω, a resonant THG occurs [1,2]. Time Δt for establishing the resonant nanoplasma frequency is determined by the average size of the clusters and their atomic composition. The subject of this work is an experimental study of the relationship between the average cluster size, time Δt and the efficiency of THG initiated by an intense femtosecond Ti:Sa by laser radiation.

The time Δt is determined by the ratio $\Delta t \approx (R/c) \times (N_0/N_{3\omega})^{1/3}$ [3], where R is the initial average radius of the clusters, N_0 is the initial plasma density, $N_{3\omega}$ is plasma density corresponding to the case $\omega_p = 3\omega$ and $c \approx 2 \times 10^7$ cm/s is the plasma speed of sound. The initial plasma density N_0 can be estimated based on the ionization state of atoms in cluster Z and its Wigner-Seitz radius r_w ($N_0 = 3Z/4\pi r_w^3$). Thus, laser irradiation in the pump-probe mode will allow us to set a delay between pulses corresponding to Δt for each average cluster radius R. Verification of the proposed method was carried out for the case of monopulse femtosecond Ti:Sa laser exposure. The pulse duration was set by changing the position of the compressor diffraction gratings. Clusters were produced by the technique based on supersonic Ar expansion (backing pressure 30 bar) through the conical nozzle (0.5 mm orifice, 5° half expansion angle) into vacuum. The exposure was carried out with focused femtosecond radiation of a Ti:Sa laser (vacuum intensity of a spectrally limited pulse of 50 fs: $I \sim 3 \times 10^{16}$ W/cm^2). The experiment is described in [4]. It is found that the maximum THG efficiency 7×10^{-5} corresponded for pulse width 300 fs (positive chirping mode). The initial plasma density was the level of 2×10^{23} cm^{-3}, corresponding on the Ar ionization state $Z = 9$ [3] and the Wigner-Seitz radius of 2.2 A. From the above formulas the average cluster radius is $R \sim 26$ nm. Estimation by the Hagena formulas [5] for the experimental conditions gives the value of the average cluster radius $R = 27$ nm, which is in agreement with the experiment within the error limits. It was obtained that the THG efficiency at pulse duration 300 fs increased linearly with cluster size (backing pressure varied in the range of 5–30 bar). The paper provides data related to the measurement of the third harmonic parameters for krypton and carbon dioxide clusters.

Thus, for the first time, the relationship between time for establishing the resonant plasma frequency and the average size of clusters was experimentally stated, which makes it possible to estimate the size of nanoparticles in laser-cluster experiments. An enhancement in the third harmonic efficiency is observed with the increase in the cluster radius.

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Ultra-broad, high-brightness coherent light source

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Hight-brightness table-top systems with multi-octave bandwidth are highly required in many applications such as molecular spectroscopy, hyperspectral spectroscopy, femtochemistry, and strong-field physics [1].

Here we present a multi-octave coherent light source with 2-5 orders of magnitude higher brightness compared to typical sycnchrotron light sources. The concept is based on a successful combination of soliton broadening and dispersive wave (DW) generation in anti-resonant reflection photonic crystal fiber (ARR-PCF) and a following intra-pulse difference frequency generation (IP-DFG) process.

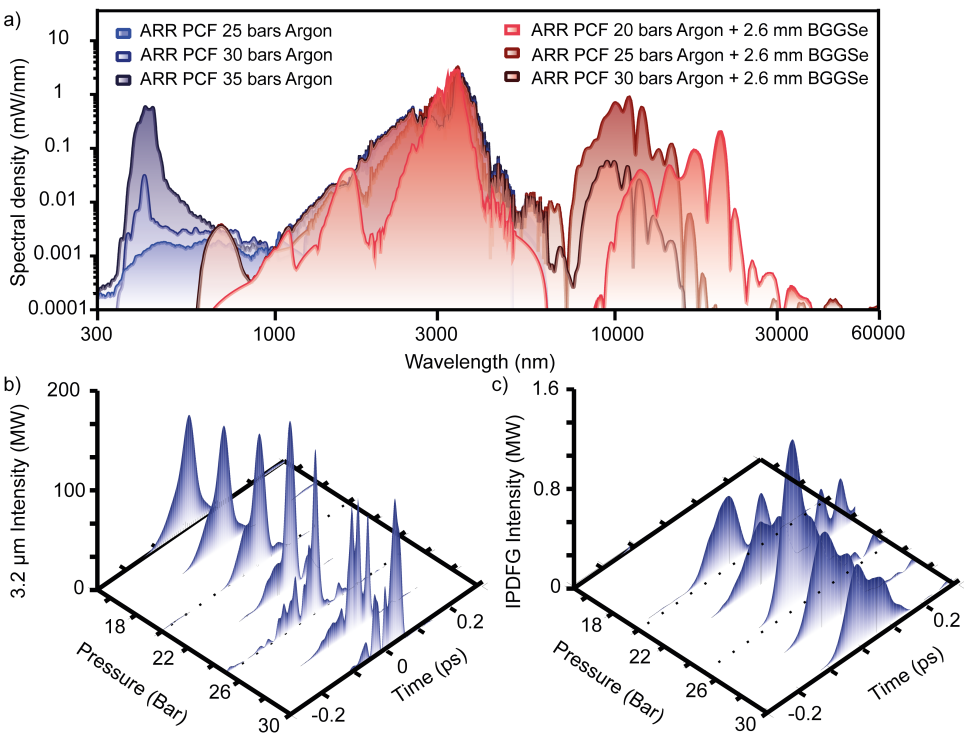

Figure 1: The overall spectrum is tunable through the Ar pressure to optimize the UV (35 bar) or the mid-IR part (20 bar). b) Evolution of the temporal pulse shape of the fiber output and c) intrapulse DFG process.

Our front-end OPCPA system provides a 3 µm, 100 fs, 160 kHz pulse train at sub-mJ level, which is lead into the ARR-PCF filled with Ar. At a gas pressure above 20 bar the UV/visible range is reached through soliton-broadening and DW generation with 2.5 MW peak power. The output is focused into a 2.7 mm thick $BaGa_2GeSe_6$ crystal [2] providing high-nonlinearity and phase-matching bandwidth to extend the spectrum through the IP-DFG process reaching

1.5 MW peak power in the mid-IR. The overall spectrum is tunable by the gas pressure inside the (ARR-PCF): at 35 bar the the UV generation os the most efficient, meanwhile at 20 bar the compressed pulse shape is optimal for the highest IP-DFG efficiency (fig. 1.a). The evolution of the temporal pulse shape (fig. 1.b) is measured by SHG-FROG demonstrating soliton compression and a temporal shock-front at 25 bar. Meanwhile the following IP-DFG pulse shape (fig.1.b.) centered at 10 µm was characterized by electro-optic sampling proving the carrier envelope stability.

Bright, coherent CEP-stable table-top source was introduced with ultra-broad spectrum covering the range from 340 nm to 40,000 nm with pulse peak-powers in the UV and THz regime of 2.5 MW and 1.8 MW respectively [1]. The advantageous characteristic indicates a plenty of important application fields of such a multi-octave system starting from nonlinear and multi- dimensional spectroscopy in the time and frequency domain through molecular spectroscopy and physical chemistry up to solid-state physics.

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Nonlinear response of 1D multi-level system calculated from nonstationary Schrödinger equation

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Theoretical description of filamentation in transparent media widely relies on nonlinear propagation equations like UPPE, NLSE, etc [1]. The nonlinear response of the medium is usually considered as independent additive responses of bound electrons (3rd and higher order polarization) and self-induced plasma calculated according to rate equations. Such a method provides only limited possibilities to account for the dispersion of nonlinearity, i.e. the dependence of nonlinear coefficients on the frequency of the optical pulse, which is important for the multi-harmonic pulses or ultraviolet pulse propagation. To provide the description of this dispersion, a quantum approach is desirable. However, full-scale $3D$ Shrödinger simulations of laser-atom interaction would be impossible to couple with $3D$ propagation simulations. For the reason of computational costs, the quantum model that can be introduced to propagation simulations must be one-dimensional at most [2]. A fair question here is how good can be $1D$ Shrödinger equation to describe the response of the realistic medium to the high-intense femtosecond laser field [3]. In this work, we develop a one-dimensional quantum model of an atom that can be introduced into propagation equations. For this system, we calculate 3rd order response as well as ionization rate in dependence on the laser pulse wavelength so as to check it for the consistency with the semi-classical models.

Since the linear and nonlinear susceptibilities of an atom are ultimately determined by the structure of its quantum levels, simple potential pits like rectangle, harmonic, or Dirac's delta potential would not be fully satisfying. We chose a Lorenz-like pit with a super-gaussian cut-off that makes the potential finite:

$$U(x) = \frac{-0.5256}{\sqrt{x^2 + 0.63^2}} \exp\left[-\left(\frac{x}{8}\right)^{16}\right] \tag{2}$$

where Hartree atomic units are used. From the stationary Shrödinger equation with the potential (1), three bound states with the energies of -12.08, -2.93 and -1.17 eV were found. The lowest two energies closely represent the ground and first exited ones of xenon atom. A third-order split-step method was used to integrate the non-stationary Shrödinger equation

$$i\partial_t \Psi = -\tfrac{1}{2}\partial_x^2 \Psi + U(x)\Psi - \mathcal{E}(t)x\Psi \tag{3}$$

with the laser field $\mathcal{E}(t) = \sqrt{I}\exp[-(t/5\text{ fs})^2]\sin(2\pi\nu t)$, which had the intensity I in range from 0.01 to 100 TW/cm^2 and frequency ν in range from 300 to 2000 THz (wavelength from 1 µm to 150 nm). The medium susceptibilities were extracted from polarization $P(t) = -\int x|\Psi(x,t)|^2 dx$.

The linear dispersion evaluated for the intensities of 0.01–1 TW/cm^2 reproduced the Sellmeier-type dependence with the resonances corresponing to the ionization potential and excited levels. The third order susceptibility estimated for intensities up to 20 TW/cm^2 after subtracting the linear response depends on the frequency with the resonant-like behavior in the vicinity of one third of ionization potential (Fig. 1, left), as supposed by the fit constructed basing on the $3D$ density functional simulations [4]. The ionization in higher frequencies clearly reproduces the multiphoton ionization [5], see Fig. 1, right panel.

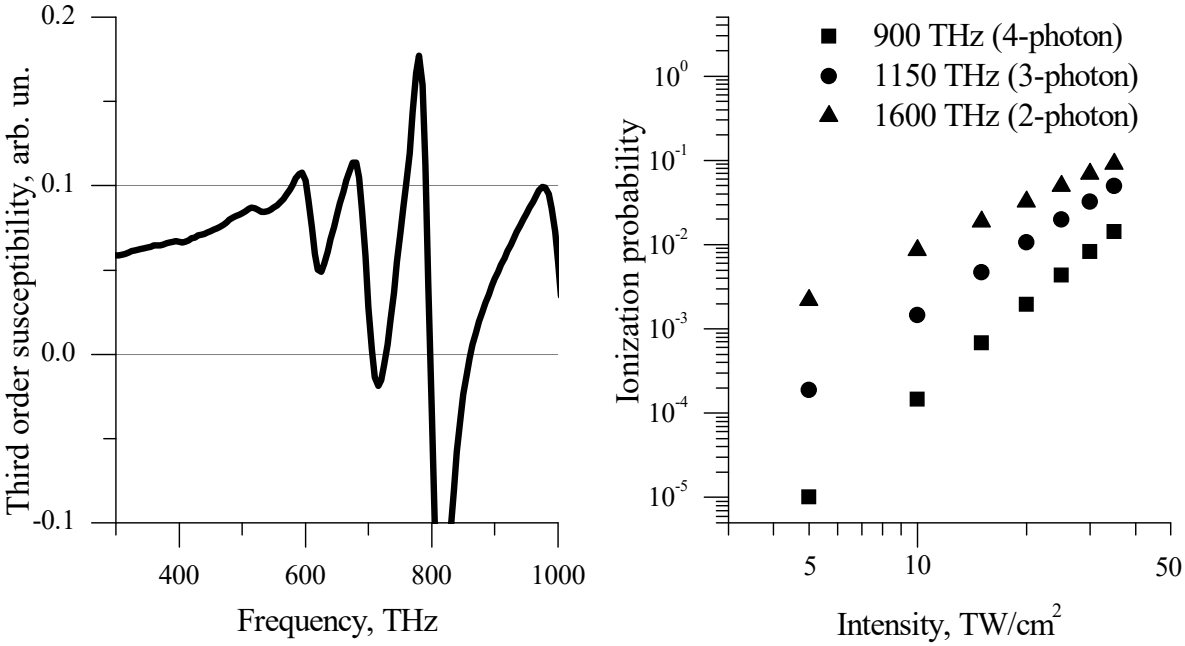

Figure 1: (Left) The dependence of third order nonlinear susceptibility on laser frequency. Note the resonance character as the frequency approaches 900 THz, which is one third of ionization potential. (Right) The dependence of ionization probability on the intensity in log-log scale for three selected frequencies representing 2- to 4-photon ionization.

This work is supported by Russian Science Foundation (21-49-00023).

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From THz air photonics to THz liquid photonics

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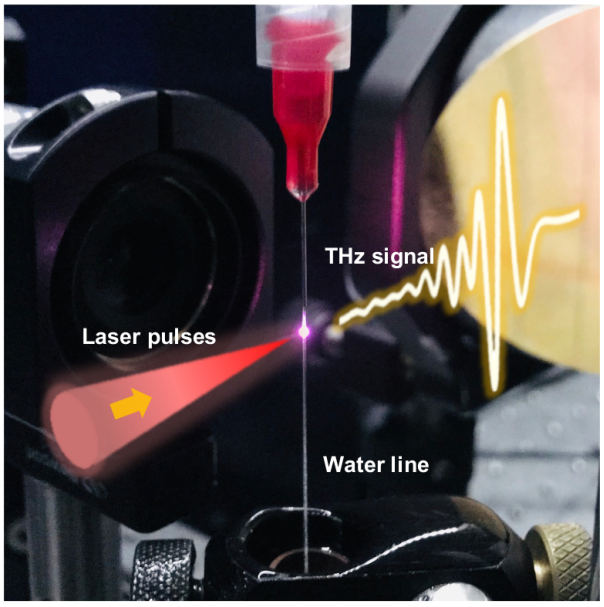
Following the steady evolution of THz technology since the late 1980s, the next frontier will possibly be the era of extreme THz science, where strong THz field-matter interactions can be investigated, and nonlinear THz spectroscopy and imaging are explored. However, fully realizing these opportunities will require the development of bright, efficient THz sources. Energetic ultrashort laser pulses are widely used to generate intense broadband THz pulses via interaction with a certain target.

Matter can exist in four distinct states in everyday life: solid, liquid, gas, and plasma. Three of the four states—solids, gases, and plasmas—have been used as sources of THz wave for decades; however, the use of liquids as THz wave emitters is extremely limited. This may be due to the high absorption of liquid water in THz range. Moreover, it is reasonable to expect that liquids should provide unique properties if they could be harnessed as THz sources. Specifically, liquids have a comparable material density to that of solids, meaning that laser pulses over a certain area will interact with three orders more molecules than an equivalent cross-section of gases. In contrast with solids, a flowing liquid allows each laser pulse to interact with a fresh area. Thus, material damage threshold is not an issue with kHz repetition rate laser pulses. This makes liquids very promising candidates for the study of high-energy-density plasma, as well as the possibility of being the next generation of intense THz sources.

We present recent progress on THz liquid photonics, which is the extension of previous study of THz air photonics. New results include low temperature liquids (nitrogen at 77 K), liquid metals, and liquids containing nano particles with their resonance at the laser excitation frequency. Figure below shows the concept of a THz wave signal generated from a waterline under short laser pulse excitation. A successful investigation on THz generation from liquids will help to search the last piece of the matter-phase puzzle for THz sources.



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Spectral broadening and phase randomization of TW-power KrF laser beam along 100-m propagation in atmosphere related to the shock ignition ICF

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Shock Ignition (SI) Inertial Confinement Fusion (ICF) with KrF laser driver is one of promising ways to achieve thermonuclear (TN) fuel ignition and energy production [1]. A smooth TN target irradiation and a broad UV laser spectrum guarantee a robust target imploding and obtaining TN energy gains ≥ 100 at laser energy ~ 0.5 MJ with a temporally profiled pulse. A multi-beam angular multiplexing scheme combined with Induced Special Incoherence (ICI) technique were implemented earlier to provide a speckle-free uniform laser light distribution and efficient energy extraction from KrF amplifiers for nanosecond pulses [2]. In the SI pulse-form peak power in the final 100–200 ps spike exceeds by 1–2 orders of magnitude the main pulse of 10–20 ns duration, that is multi-TW power should be transported along ~ 100 m air pass of the multiplexer.

In the present experiments at GARPUN-MTW Ti:Sa/KrF laser facility on the propagation of TW-power, 1-ps UV laser pulses along 100-m air pass we have observed two nonlinear optical effects important for the SI ICF, namely (i) spectrum broadening due to Rotational Stimulated Raman Scattering (RSRS) in atmospheric N_2 and (ii) a break-off of multiple filamentation with phase randomization of a coherent supercritical laser beam (with peak power 3–4 orders of magnitude exceeding critical power for Kerr self-focusing $P_{cr} = 0.1$ GW) at distances over 60 m caused by self-phase modulation (fig. 1) [3]. An unfavorable filamentation appearance between amplifier stages was shown to be suspended by using a cell with Xe, which de-focuses filaments due to a large negative resonance-enhanced nonlinear refraction index.

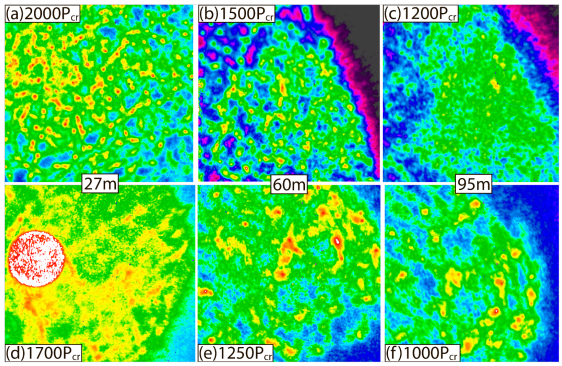

Figure 1: Beam patterns at various distances from the final KrF amplifier in a chain: (a–c) without and (d–f) with a Xe cell. Panel size is 4×4 cm. Peak power in the units of P_{cr} is shown in inserts.

We acknowledge the contribution of A. A. Ionin, D. V. Mokrousova, L. V. Seleznev and E. S. Sunchugasheva in the experiments. The work was supported by RFBR grant No. 19-02-00875.

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Section 5: Frequency combs in spectroscopy and optical clocks

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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

Time and frequency transfer

Optical frequency combs applications

Quantum metrology with weakly coupled atomic condensate solitons

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Nowadays rapidly growing experimental facilities in quantum technologies evoke an increasing interest in quantum metrology and sensing [1] which possess many important applications, such as verification of fundamental physical laws with gravitation, frequency standards and atomic clocks, magnetometers, quantum gyroscopes, etc. Traditionally, such schemes exploit Mach-Zehnder interferometers (MZI) based on ultracold atomic ensembles and Bose-Einstein condensates (BEC). In this work we discuss condensate quantum bright solitons for achieving ultimate unknown phase parameter (ϕ) estimation [2]. We assume that MZI arms accumulate some (relative) phase proportional to ϕN^k, where N is particle number, k is a positive integer number. The best precision of the measurement (without losses), $\delta\phi_{min} = \frac{1}{N^k}$, can be obtained with an ideal balanced maximally path-entangled two-mode $N00N$-state. We have shown that the BEC solitons provide unprecedented accuracy for phase estimation among current Kerr-like systems within nonlinear quantum metrology approach, possessing $k = 3$ [3,4].

At first, we suggest an atomic soliton Josephson junction (SJJ) device for initial state preparation purposes. SJJ consists of two weakly-coupled condensates with negative scattering length. The condensates are trapped in a double-well potential and elongated in one dimension. We comprehensively studied superfluid – Mott insulator like transition in such a system that occurs at some effective parameter Λ_c that accounts condensate particles interaction and their inter-well tunneling. For $\Lambda \geq \Lambda_c$ this transition led to formation of entangled Fock state, revealing dominant $N00N$-state components and exhibiting practically important resistance to weak particle losses [3,4].

In the presence of particle losses we model coupling of MZI with environment by means of the fictitious "beam splitter" approach that characterized effectively by transmissivity parameter η. We have shown that standard interferometic limit in general case may by described as $\delta\phi_{SIL} \propto 1/\sqrt{\eta}N^{k-1/2}$. The value of $\delta\phi_{min}$ for our scheme depends on Λ-parameter, which is relevant to the performance of the SJJs as a device for probe state preparation. At moderate values of η the phase estimation accuracy approaches standard interferometric limit $\delta\phi_{SIL}$ for the SJJ system close to the superfluid regime, $\Lambda < \Lambda_c$. The behaviour of $\delta\phi_{min}$ beyond critical point $\Lambda \geq \Lambda_c$ is determined by the properties of entangled Fock states at the input of MZI in the presence of losses. We have shown that operating near the critical point Λ_c we can achieve essential improvement of accuracy of the phase estimation even in the presence of particle losses. In particular, for weak losses operating close to the Mott insulator regime the accuracies of the linear (k = 1) and nonlinear (k = 3) metrologies attain the Heisenberg ($\delta\phi_{min} = 1/N$) and super-Heisenberg ($\delta\phi_{min} = 1/N^3$) limits, respectively. We also discuss feasibility of experimental observation of predicted effects in the framework of current experiments performed with atomic BEC bright solitons [5].

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Progress of silicon cavities with crystalline mirrors for infrared laser stabilization

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Ultrastable laser systems find wide application areas such as gravitational wave detection [1], dark matter search [2], and fundamental constants drift investigation [3]. Optical clocks need such laser systems. The fractional instability of the best modern optical clocks reaches the level of $6 \cdot 10^{-19}$ at averaging time of one hour with the help of a laser system stabilized to a cryogenic monocrystalline silicon reference cavity [4].

Here we report on a pair of identical laser systems based on silicon cavities with crystalline coatings operating at 1550 nm. To suppress frequency instability we placed silicon cavities in vacuum cryostats and cooled them down to 124K, which corresponds to zero thermal expansion point of silicon. We measured the finesses of both cavities which appeared to be 200000 and 143000. Each eigenmode of both cavities is split into two components corresponding to different light polarization with a frequency separation of 150kHz, which means that the crystalline mirrors coatings are birefringent. Features of silicon reference cavities, crystalline supermirrors, 1550 nm fiber lasers frequency stabilization will be discussed. Preliminary results of frequency comparison of the laser systems will be presented.

This work is supported by the Russian Science Foundation (Grant 19-72-10166).

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Methane photonic microwave oscillator - experience of testing at long-term operation

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Compact and reliable low phase noise photonic microwave oscillators (PMO) with short-term stability (5-1)$\times 10^{-15}$ for average time $\tau = 1$ s are needed for variety of applications – frequency etalons based on atomic fountains, radio astronomy, radars, etc. Common approach for their development is based on stabilization of semiconductor laser frequency over high-Q Fabry-Perot resonator and further optical-to-microwave frequency division [1, 2]. To divide the optical frequency to the microwave range a femtosecond system based on a fiber laser is usually used. After two decades of research and development these systems are close to step from laboratory environment to practical applications. Our research has a similar goal in development of a highly stable PMO but as a source of stable optical frequency the He-Ne/CH_4 optical frequency standard ($\lambda = 3,39$ µm) is chosen [3].

At present two PMO based on a femtosecond Er fiber laser optical to microwave divider and He-Ne/CH_4 optical frequency standard have been created and tested over long periods of operation. Results of Allan deviation and phase noise spectral density measurements for the created PMO as well as reliability of operation will be presented and discussed at the Conference.

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Repetition frequency stability of mode-locked Cr:ZnSe laser

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Nowadays, compact stable radiofrequency synthesizers based on an optical frequency standard (RS-OFS) have proven their demand by applications in the time-frequency metrology, navigation or RS-OFS can be used as simple laboratory tools delivering stable frequency. Several realizations of the RF-OFSs based on the utilization of a mode-locked laser and an ultrastable CW laser with frequency instability in the range from 10^{-15} to 10^{-18} have been demonstrated already [1, 2]. Both key elements of the RF-OFS (a stable CW laser and a mode-locked laser) can be designed in different ways with various benefits as well as drawbacks [3].

One of the stable CW laser concepts is based on the laser frequency stabilization over narrow absorption lines of gases. A concept of stabilized CW Cr-doped zinc selenide laser to absorption lines of methane at the wavelength of 2.36 µm was already demonstrated in [4], and the same active crystal can be effectively used for building of mode-locked lasers with a repetition rate of up to 300 MHz [5], which can be easily optically locked to the stable CW Cr:ZnSe laser due to intrinsic overlap of the lasers operation wavelengths.

In this study, we measure repetition frequency stability of a free-running SESAM mode-locked Cr:ZnSe laser that can operate in a single pulse or a bound state regimes with 130 MHz repletion rate [6].

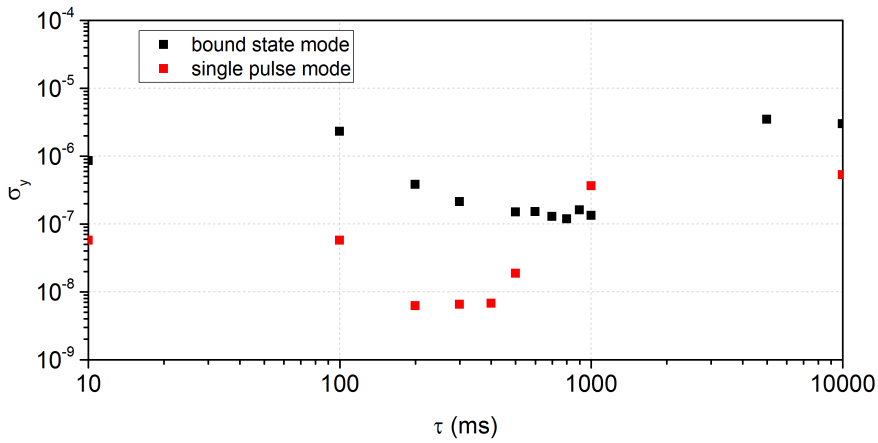

Figure 1: Relative Allan deviation for the free-running operation.

The relative Allan deviation (Fig. 1) is measured over the averaging time interval from 10 ms up to 10 s. For the bound state operation regime, the Allan deviation at the averaging time of 1 s is $1.3 \cdot 10^{-7}$, that is slightly lower than for the single pulse mode. On the other hand, the relative Allan deviation at short-term averaging times demonstrates smaller values for the single pulse operation.

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Second-order Zeeman shift suppression in thulium optical clock using synthetic frequency

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Optical clocks have a lot of applications in fundamental and applied science, some of which require transportable systems [1]. Neutral thulium has unique properties that make it a prospective platform for transportable optical clocks. A 1.14 µm clock transition in thulium has low sensitivity to external static electric fields and blackbody radiation of the environment [2] which is the main problem of many other atomic systems. Other effects that cause frequency shifts in thulium optical clock were discussed in [2], with the most significant one being Zeeman effect.

To avoid frequency shift assosiated with linear Zeeman effect, we use the transition between central magnetic sublevels of ground and clock states ($|m_F = 0\rangle \rightarrow |m'_F = 0\rangle$). To eliminate the second-order Zeeman shift, we plan to use so-called synthetic frequency technique. In this approach we are interrogating both hyperfine components of the clock transition. These components have the same absolute value of second-order Zeeman shift but with different signs. Because of that, second-order Zeeman shift for the mean "synthetic" frequency of the two clock transitions between hyperfine components cancels out (see fig. 1).

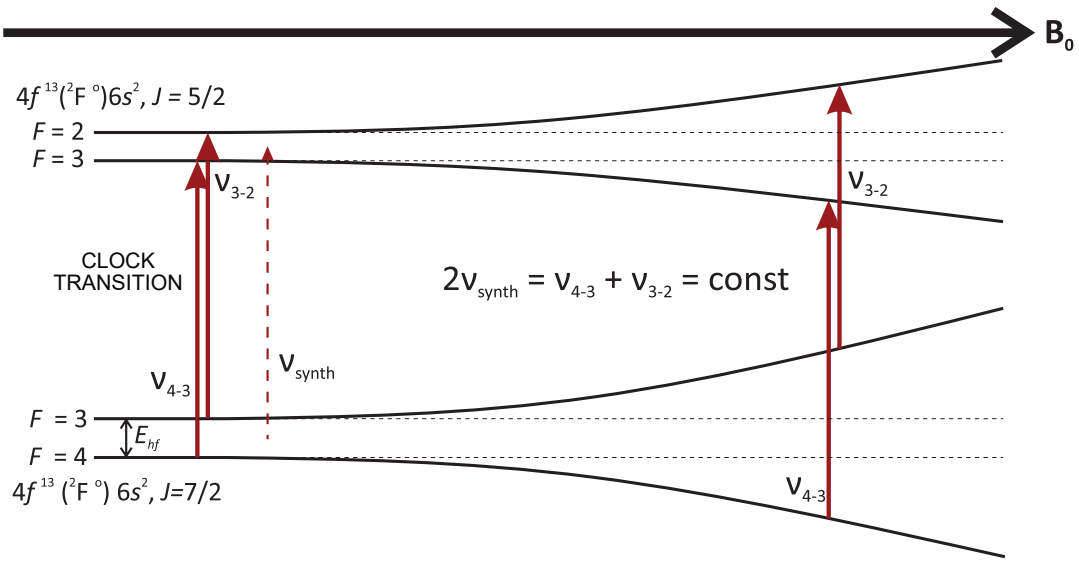

Figure 1: Concept of synthetic wavelength

In this work we experimentally realized digital locking of the laser to both clock transitions and demonstrated the suppression of the second-order Zeeman shift for the synthetic frequency.

The work was supported by the Russian Science Foundation (Grant No. 19-12-00137).

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Analysis of optimal soliton pulse duration for quantum noise squeezing in optical fiber

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Quantum noise suppression of light is desirable for many applications including detection of gravitational waves, quantum sensing, and quantum communications [1]. There are several ways to obtain continuous wave (CW) or pulsed squeezed light. The potential for achieving squeezing in optical fibers is not yet fully exploited, and possibilities of efficient noise reduction in fibers are studied. For quantum noise suppression of CW radiation, fiber lengths of about a hundred meters are required, and limiting factors are optical loss and guided acoustic wave Brillouin scattering (GAWBS). For solitons with duration ∼0.1–0.2 ps and a high peak power, significantly shorter fibers are required (of about ten meters), and instead of loss or GAWBS, Raman effects limit squeezing.

Here we analyse the possibility of quantum noise squeezing of solitons with durations of 0.1 -– 1 ps in an optical fiber with an enlarged mode area with realistic parameters using home-made software. The simulation of pulse propagation with allowance for quantum noise entails modelling multimode many-body quantum system dynamics. We achieve this here with a truncated Wigner technique, which provides accurate results for relatively short propagation distances and large photon number. We model the Raman-modified stochastic nonlinear Schrödinger equation [2] with allowance for losses. GAWBS is neglected because its influence scales down with increasing effective mode field area. The initial condition is a fundamental soliton with an addition of normally distributed stochastic quantum noise. In modelling, we switch on/off Raman effects and loss to study the interplay between limiting factors for solitons with different durations.

For fundamental solitons, the peak power is inversely proportional to the square of the pulse duration, so for longer pulses with duration ¿0.6 ps the required distances are longer and the loss impact is stronger. For those pulses, optical losses limit squeezing and Raman effects are insignificant. For short solitons with duration < 0.2 ps, the Raman effects limit squeezing because their spectra are wider, so the overlap with spectrum of the Raman function is larger. The results of our model suggest that there is an optimal soliton duration ($T_{FWHM} \sim 0.4$ ps) when the balance between these limiting factors is satisfied and the strongest squeezing is better than -22 dB.

This work was funded by the Mega-grant of the Ministry of Science and Higher Education of the Russian Federation, Contract No.14.W03.31.0032 and by the Russian Foundation for Basic Research, Grant No. 19-29-11032.

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Atmospheric short link for the ultra-stable optical frequency signal transfer

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Coherent transfer of frequency and time signals over countries and continents, between Earth stations and satellites is rapidly developing area of research and technology. A network of optical clocks connected with links [1] create great opportunities in such fields of science and technology as the formation of national and international time scales, relativistic geodesy, satellite navigation, very-long-baseline interferometry, tests of fundamental theories, search for dark matter. The best modern optical clocks have reached [2,3] the level of relative uncertainty and instability of 10^{-18}. Transferring signals of such kind of standards without losing of their characteristics using radio frequency methods is impossible, since the latter cannot provide [4] fractional instability better than 10^{-16}. However, it is possible to reduce the level of the phase noise introduced by the communication link by transferring signals at optical frequencies and using active noise cancellation technique. The rapid development of stationary and transportable [5] optical clocks shows the necessity of designing both fiber [6] and open-air [7] links for the highly stable signals transfer.

Here we report on the developing an open-air 17 m link operating at 1550 nm. We have obtained more than 11 000 s of uninterrupted data and have showed that our active noise compensation system allows to reduce the fractional frequency instability induced by the link from $2.6 \cdot 10^{-16}$ to $1.7 \cdot 10^{-19}$ at averaging time $\tau = 1000$ s in terms of Allan deviation calculated from data measured by K+K phase recorder. The link contribution to inaccuracy of transferred signal is reduced from $1.9 \cdot 10^{-17}$ to $5 \cdot 10^{-20}$.

In continuation of this study we plan to increase the link length up to 500 m and use an unmanned aerial vehicle with a mirror fixed on it as a moving receiver model.

This work is supported by the Russian Science Foundation (Grant 19-72-10166).

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Section 6: Physics and technology of ultrafast lasers and ultrashort laser pulses

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Scope

Solid-state, parametric, fiber, and hybrid laser systems

Stretchers, compressors, and phase control

Measurement and characterization of ultrashort pulses

Laser design and related issues
Innovative femtosecond technologies

Raman and laser generation in silica, chalcogenide, and tellurite glass microresonators in the telecommunication C–, L–, and U–bands

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Microresonators with whispering gallery modes (WGMs) are of great interest for a lot of applications [1]. Due to high Q-factors and strong localization of the WGM field in a small volume, high light intensities are achieved at low pump powers, which results in very low thresholds for nonlinear and laser effects [1,2]. WGM resonators are made of various materials, with a constant search for new solutions that have advantages either in technological terms or in terms of expanding the boundaries of the observed physical effects [2]. Here we focus on Raman generation in silica and chalcogenide glass microspheres and laser generation in Er-doped tellurite glass microspheres.

We demonstrate the single-mode Raman generation widely tunable in the U–band and beyond in a silica microresonator. The Stokes wavelength is tuned in the $1.631 - 1.685$ µm range when tuning a C–band pump laser in the $1.520-1.570$ µm range [3]. We found switching between Raman modes which corresponded to jumps by 2 or 3 WGMs while sweeping the pump laser near a certain WGM (without pump jumps between WGMs) [3] and explained this effect theoretically.

Next, in an As_2S_3 chalcogenide glass microsphere we achieved experimentally the single-mode Raman generation tunable in the L–band and the U–band from 1.610 µm to 1.663 µm by tuning the pump wavelength in the $1.522 - 1.574$ µm range [4]. The experimental results are in a good agreement with numerical results based on the Lugiato-Lefever equation modeling [4].

We also report experimental and theoretical results on the laser generation in an Er-doped tellurite glass microsphere under in-band pumping. Depending on the system settings, single-mode and multimode lasing was attained experimentally in the C–band near 1.56 µm and in the L–band near 1.60 µm, as well as simultaneously in both bands [5]. To describe lasing, we use the original analytical model with allowance for the mode competition and verification of the stability of found solution [5]. This is necessary for better understanding of the observed regimes and further control and optimization of such systems. We found theoretically that for low Q-factors lasing occurs in the C–band but for relatively high Q-factors lasing occurs in the L–band, which agrees with the experimental observations. Switching between the generated wavelengths from the C–band to the L–band occurs with a jump which was observed experimentally and explained theoretically [5].

The obtained results can speed-up progress in the development of microresonator-based devices operating in the telecom range and give a better understanding of their theoretical aspects.

This work is supported in part by the RSF, Grant № 20-72-10188 and in part by the Mega-grant of the Ministry of Science and Higher Education of the RF, Contract № 14.W03.31.0032.

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Spectral-temporal patterned supercontinuum generation at a multi-gigahertz repetition rate

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The supercontinuum (SC) is a key technology for many applications ranging from ultra-precise metrology and spectroscopy to biomedical studies and sensing. In most works the SC generation is achieved in highly nonlinear devices (e.g., nonlinear fibers) pumped by a train of ultrashort pulses. In this case, the SC is a train of more or less the same pulses with broadened spectrum. We present the generation of SC, which is a train of ultrashort pulse bunches with a spectrum controllable from pulse to pulse. Moreover, the distance between the pulses is adjustable in a broad range from 5 ps to several nanoseconds, resulting in an effective repetition rate of up to 200 GHz. The experimental system is based on a specially designed Er-doped mode-locked fiber laser that generates bunches of pulses in which the distance between the pulses can be adjusted by tuning an intracavity Mach-Zehnder interferometer [1]. The laser supports two types of pulses circulating in the cavity: pulses with high and low amplitude (H- and L-pulses), which can be assembled into patterned bunches. Bunches with a large number (up to 250) of identical pulses and bunches with interleaved H- and L-pulses can be generated. The laser output is amplified in a fiber amplifier up to 400 mW of average power and used as a seed for SC generation in a highly-nonlinear dispersion shifted silica fiber. The spectral broadening of H- and L-pulses is different, so that a patterned spectral-temporal structure is formed. The integrated SC spectrum spans from 1.35 µm to 1.8 µm. The time-resolved spectrum is measured by a monochromator with a fast photodetector, and the temporal structure is confirmed by ultrafast all-optical sampling. By selecting spectral windows with appropriate central wavelengths, different temporal signal structures can be generated. The developed source can be used for the generation of high-speed test patterns and reference signals for ultrafast optical waveform synthesis, optical signal reconstruction, and ultrafast spectroscopy.

This study was supported by the Ministry of Science and Higher Education of the Russian Federation (Contract № 14.W03.31.0032).

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Ultra-high peak and average laser systems (problems and solutions)

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In a new generation of ultra-high peak and average power laser systems the relatively high pulse energy should be combined with the high repetition rate and connected with it a thermal management. Ti:Sa amplifiers with crystals of the different geometries can satisfy this request for different output parameters. Benefits of using the active medium of a thin disk (TD) [1-3] and slab configurations are thoroughly evaluated and tested numerically and experimentally. The simulations revealed the existing limitations for heat extraction in TD geometry in the sub-Joule energy regime for higher repetition rate operation. Geometry conversion from TD to a thin slab (TS) and cross thin slab (XTS) arrangement [4] significantly increase the cooling efficiency with an acceptable crystal temperature for pump average power values up to few kW with room temperature cooling, and up to tens of kW with cryogenic cooling. Direct diode pumping simulated for CW regime have demonstrated 1.4 kW output power with 34% extraction efficiency using room temperature cooling and more than 10 kW and $\sim$ 40% efficiency with cryogenic cooling. These ideas were used in CPA laser systems. One of the first such systems is under the development and investigation in Colorado State University. Very important element for that, pump laser, was developed there and will be also presented in the talk. It was demonstrated the generation of 1.3 J pulses of few nano-second duration at 1 kHz repetition rate from a diode pumped Yb:YAG laser. The laser employs cryogenically cooled amplifiers to generate $\lambda = 1030$ nm pulses, that were converted to the second harmonic of 515 nm with efficiency up to 80%, which allowed to achieve the 1 J pulse energy of this wavelength [5]. This laser has been used as a pump of Ti:Sa amplifiers of the discussed geometry for generation the ultrashort femtosecond pulses of sub-joule energy and 1 kHz repetition rate. The recent progress of these experiments will be presented. Solutions of the pump beams delivery and thermolens compensation will be also considered.

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

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Laser induced forward transfer (LIFT), is a high resolution and fast printing technique, which has proved its efficiency in various applications that require micro resolution precisions. Among numerous varieties of the method, a blister-based-laser-induced forward transfer (BB-LIFT) is a technique that allows soft and clean transfer of materials, particularly attractive for biomolecules and gentle nanomaterials [1,2]. In this approach, a thin metal film is used as an intermediate absorbing layer of laser pulse to generate a transient mechanical deformation (blister) and thus to softly remove materials located on the film.

We have developed a BB-LIFT set up which allows visualization of the printing process from both donor and receiver substrates; hence, the precision and amount of material transfer can be monitored and adjusted in-situ. In this work, we investigated resolution of the printing system through a study of the laser spot size effect on blister formation for picosecond pulses of a Yb:KGW laser (1030 nm, 6 ps). A glass substrate coated with Si (5 nm) - Ni (200 nm) - Ti (5 nm) layers, was used as a test sample. The laser spot size was varied in the range of 5–100 µm using different focusing optics. For each focusing conditions, blisters were generated in a relatively narrow fluence range within the scope of the spot size (typical examples are shown in Fig. 1). The blisters morphology was characterized by optical and atomic force microscopies. The blister formation mechanisms are discussed.

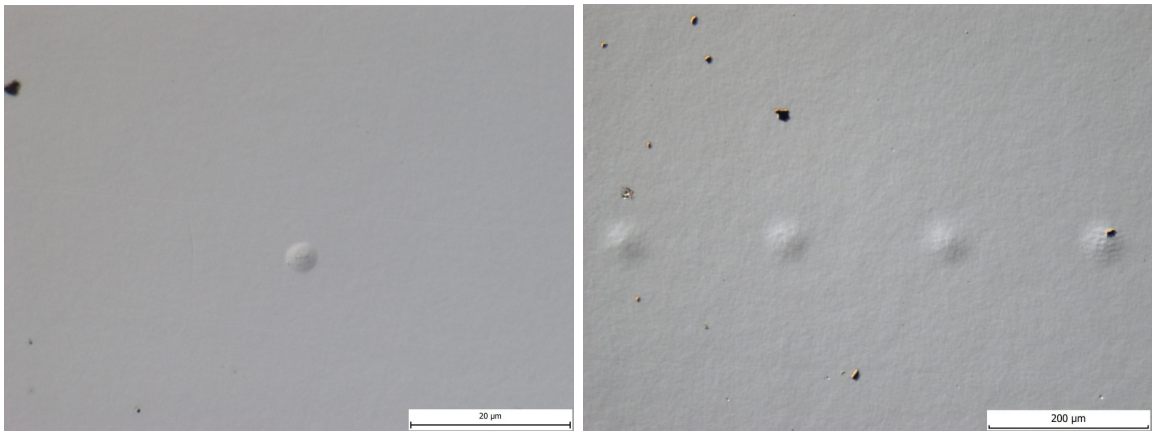

Figure 1: Optical images of blisters of 4 and 100 µm generated with single ps laser pulses

The experimentally obtained frequency shifts of the Stokes intracavity SLFRS components correspond to those calculated values which were obtained using the Lamb approach, taking into account the influence of the environment.

It is experimentally shown that intracavity SLFRS has a much higher potential, allowing one to obtain components of a higher order than in conventional SLFRS.

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Generation of femtosecond pulses from the radiation of a sub-ps ytterbium pump laser

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The development of laser radiation sources with high peak power has become one of the most promising areas in laser physics. In this regard, much attention has been paid to research in the field of ultrashort pulses. As a result, a new direction began to develop – the interaction of ultrashort pulses of powerful coherent radiation with matter. Using a femtosecond laser, it is possible to create and investigate unusual extreme states in a solid.

The work was carried out on the creation of multi-TW laser radiation sources. Using a new approach to generating femtosecond pulses directly from the radiation of a pump laser, a parametric system was developed and manufactured that generates femtosecond pulses with an energy level of tens of µJ and a pulse duration of several field oscillations in the near and middle infrared ranges.

To date, a traditional method is used for such purposes, that is, a high-intensity femtosecond titanium-sapphire laser is used to pump a parametric amplifier. However, this approach is not optimal, since it is possible to similarly use ytterbium picosecond lasers, which have a high pulse energy at a repetition rate of tens and hundreds of hertz and are much cheaper and easier to use [1].

The sub-ps pulses of the ytterbium laser are partially converted to the 2nd harmonic and are used to generate supercontinuum radiation. This broadband radiation is parametrically amplified.

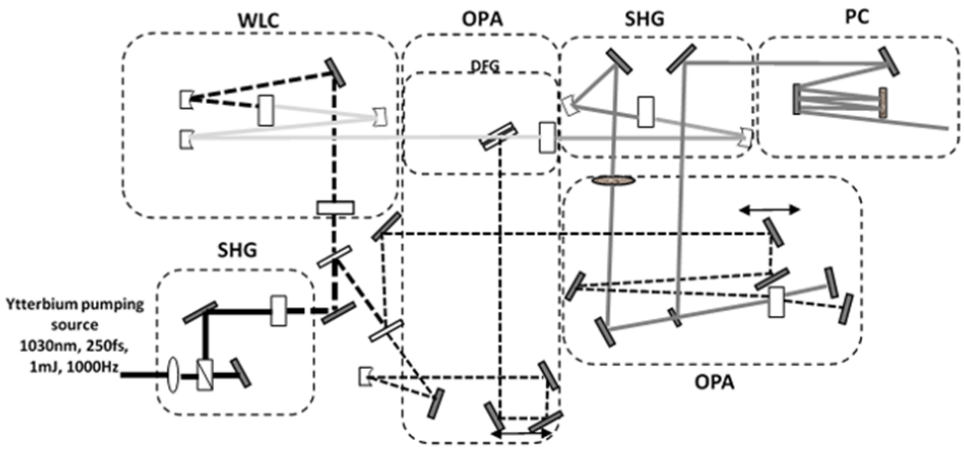

Figure 1: Laser system diagram

As a result, “idle” radiation is generated at a difference wavelength, which has the property of stabilizing the phase of the e/m field relative to the envelope (CEP) [2]. The “idle” wave entering the parametric amplification node can be additionally used using the source radiation as a pump.

Alternatively, the difference frequency radiation can be converted to the 2nd harmonic and also amplified by the 2nd harmonic of the original radiation. This generated broadband radiation can be compensated to a spectrally limited duration. To study the spectral and temporal characteristics, laser pulses were studied using a device using the FROG-SH method.

An important advantage of the developed design for generating fs laser pulses is their optical synchronization with the pump laser pulses of high-power parametric systems. Based on the results obtained, 2 femtosecond control systems with a central wavelength of 910 nm and 2100 nm with a kilohertz pulse repetition rate were manufactured. In the mid-IR laser system, more than 20 µJ of pulse energy was achieved with a duration of 35 fs. Further, this signal will be further parametrically amplified to the multi-TW peak power level. The installation with near-infrared radiation reached 15 µJ with a duration of 27 fs, it will be used as a starting point for the P of the PEARL laser complex.

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Coherent combining of radiation transferred through a multicore fiber with 25 coupled cores

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Multicore fibers are promising for increasing average and peak power in fiber laser systems. Due to tight localization of light in a fiber, the peak power of radiation that can be transferred through a single-core fiber is limited by non-linear effects. In particular, self-focusing limits the peak power of a pulse even in large-mode-area fibers. Using multicore fibers (MCF), it is possible to raise the peak power limit up to N times, where N is the number of cores. However, in order to use radiation transferred through an MCF, it needs to be combined in a single beam, and a special optical system is required for that.

In the case of non-interacting cores in MCF, the combining system must include an active feedback system that compensates for phase fluctuations between the cores. Moreover, known optical schemes for coherent combining usually require special optical components, and their complexity scales with the number of cores. Contrary to that, in an MCF with coupled cores light propagates in coherent supermodes, so no active beam phasing is required. Also, the fill factor in MCFs with coupled cores is much higher, and it becomes possible to coherently combine the radiation with the use of a few off-the-shelf optical elements.

We experimentally demonstrated coherent combining of the out-of-phase supermode transferred through an MCF with a 5×5 square array of coupled cores. The MCF supports 25 supermodes, but the out-of-phase supermode having interleaved $0/\pi$ phases in neighboring cores is the most promising supermode for power scaling because it is stable at high power [1]. We created a scheme that allowed us to control the content of different supermodes in an MCF. We used this system to launch 50-ps chirped pulses at 1030 nm into the MCF with 90% of the power contained in the out-of-phase supermode. We combined the light radiating from the MCF using the out-of-phase regime of the tiled aperture combining method. Because the cores were tightly packed, lens array and phase correctors were not required, and four coherently combined beams were formed in the far field according to [2]. We combined the 4 resulting beams using two regular 50/50 beamsplitters, achieving a total efficiency of 60%, which was limited by imperfect structure of the cores in our MCF, non-ideal excitation of the-out-of-phase supermode, losses at the beamsplitters, and some depolarization. The M^2 of the combined beam was 1.3. The system didn't require any phase stabilization to maintain high combining efficiency.

We measured the intensity and phase distributions of the out-of-phase supermode and studied them in numerical modeling. We found that in the ideal case this supermode can be coherently combined in a single beam with 93% efficiency. The M^2 of the modeled combined beam was 1.1.

This study was supported by the Ministry of Science and Higher Education of the Russian Federation (Contract 14.W03.31.0032).

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Stimulated Raman scattering of chirped Ti: Sapphire laser pulses in $BaWO_4$ crystal

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Previously we have demonstrated high efficient transient stimulated Raman scattering (SRS) of femtosecond visible-range (0.475 µm [1] and 0.515 µm [2] wavelengths) laser pulses in $BaWO_4$ crystal. In this work we study SRS in $BaWO_4$ crystal pumped by near-IR laser pulses of femtosecond Ti:Sapphire lasers with wavelengths of 0.75 µm and 0.92 µm. Since a SRS gain for emission with longer wavelength is less, an achieving of SRS threshold requires higher emission intensity that results in other undesirable nonlinear optical effects such as self-phase modulation and self-focusing. To prevent these undesirable effects, experiments were carried out with chirped laser pulses stretched to picoseconds durations.

The first experiment was carried out with Ti:Sapphire laser pulse stretched to 200 ps (the transform limited duration of 90 fs) with wavelengths of 0.75 µm. A feature of this experiment was a broadband nanosecond seeding of the SRS conversion. The SRS threshold ($\sim$ 1% conversion efficiency) was obtained at SRS exponential gain $G \approx 15$ due to the SRS seeding. At laser intensity increasing the SRS efficiency grow up to 3%, however, addition SRS peaks appeared and pump laser pulse spectrum became broader. These effects disturbed the SRS efficiency enhancement. An addition of a second $BaWO_4$ crystal to the optical scheme allowed us to increase the SRS efficiency up to 10%. Also the experiment with Ti:Sapphire laser pulses stretched to 70 ps (the transform limited duration of 300 fs) with wavelength of 0.92 µm was carried out. In this experiment without a broadband nanosecond seeding the SRS efficiency was up to $\sim$ 30%.

Thus we experimentally demonstrated the relatively high SRS efficiency (10% and 30% for emission wavelengths of 0.75 µm and 0.92 µm, respectively) for femtosecond Ti:Sapphire laser pulses stretched to picoseconds durations.

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Raman modes excitation of $BaWO_4$ crystal under traveling high-intensity 515-nm 300-fs laser pulse

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Frequency conversion of laser radiation via stimulated Raman scattering (SRS) is one of the major techniques to extend the applications of well-developed laser systems. For ultrashort laser pulses (with duration shorter than the dephasing time of SRS medium) SRS goes in a transient regime with significantly lower efficiency as compared to steady-state SRS [1]. An increase in a SRS gain increment by an increase in laser pulse intensity leads to an appearance of other nonlinear processes, e.g. self-phase modulation (SPM). The influence of SPM on SRS conversion efficiency is considered in a number of works [2-4], and negative effect is usually noted [3,4].

In this work, frequency conversion of ultrashort laser pulse in a Raman-active crystal $BaWO_4$ (BWO) was investigated under concurrent SPM and SRS effects. The second harmonic laser pulse of the Satsuma ytterbium fiber laser (Amplitude Systems) had the following properties: duration of 300 fs (FWHM), wavelength of 515 nm, repetition rate of 1 kHz, and energy up to 3.4 µJ, which was varied by detuning second harmonic crystal. This radiation was tightly focused (f=35 mm) into BWO crystal with a thickness of 8 mm. The crystal c-axis was perpendicular to the polarization of the radiation. After the crystal, radiation was focused onto a scattering plate installed in front of the entrance slit of the spectrometer (Avesta-150 FT).

A laser pulse energy increase led to a broadening of the spectrum towards a red wing and the formation of spikes structure of the spectrum. The specific spectral peaks were detected which appear sequentially, increase to a maximum, and decay. These peaks correspond to frequency shifts of 320±20 cm^{-1}, 465±20 cm^{-1}, 600±20 cm^{-1}, 830±20 cm^{-1}, 920±20 cm^{-1}. Some of these components are in a good agreement with lines of the spontaneous Raman spectrum [5]: 332 cm^{-1}, 346 cm^{-1}, 831 cm^{-1}, 925 cm^{-1}. The 465 cm^{-1} line corresponds to the linear combination of 345 cm^{-1} and 102 cm^{-1} Raman modes, the 600 cm^{-1} line can be related with overtone vibration excitation of 332 cm^{-1} mode.

Thus in this work we experimentally demonstrated the interplay of concurrent SPM and SRS effects in $BaWO_4$ crystal that resulting in spectral red-shift of laser pulse which is similar to soliton self-frequency shift observing in optical fibers [6].

The work was supported by Russian Foundation for Basic Research (20-32-70015).

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THz wave generation in Al and O doped GaSe

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It is well known the physical properties of the GaSe can be significantly modified by a doping. We proposed Al-doping to modify its resistivity, free carriers' mobility, and live-time for application as THz generator. The GaSe:Al single crystals were grown from various GaSe:Al melts by modified Bridgman method. Thin samples were careful cleaved from the ingots. As a result, no treatment or polishing of surfaces was not necessary. Numerous optically clear samples were selected for the study. Incorporation of small contents of alumina provided higher resistivity (up to 107 Ohm· cm), which is necessary, for example, to increase bias voltage or decrease absorption coefficient. The best samples containing about 0.05 mass% of Al show 20% lower lifetime and 5 times higher (up to 600 cm^2/V sec) free carriers mobility compared to that of pure GaSe. Spectral response of GaSe:Al dipole antennas at wide spectral range 0.25-4 THz was about 35 times to that of pure GaSe.

Additional doping with O was also carried out. On the initial low level doping noticeable decrease of optical loss coefficient is fixed as well as strong increase of hardness. Further doping over 0.01 at.% results in rapid growth up of the absorption coefficient. This process is followed by fast increase of ordinary wave refractive index. No changes of nonlinear susceptibility were observed at this level doping. So, the Al doping is the efficient way to strengthen mechanical properties of GaSe and improve exploitation parameters but doping with O allows extra efficient control of phase matching conditions.

For the experiential study of THz wave generation an exceedingly high optical quality GaSe specimens were selected for the test. After averaging of numerous test results of different samples, it was established evident dependance of the THz generation efficiency on the specimen thickness: the thinner specimen the lower absorption coefficient and higher efficiency normalized to the sample thickness. Finally, it was found nonlinear coefficient to be as high as from 70 to 75 pm/V versus commonly known value of 54 pm/V [1]. So, the real nonlinear coefficient is masking by the accumulating defects.

This work was supported by the Ministry of Science and Higher Education of the Russia Federation № 121031300155-8.

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Four-wave mixing in silica microresonators with germanosilicate coating

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Optical microresonators with a characteristic size of hundreds of micrometers are widely used in fundamental research and applications. They are primarily used for the generation of optical frequency combs, laser generation, spectroscopy, optical filtering, and so on [1]. Microresonators may be of different shape and material, thus can have a wide variety of dispersion and nonlinear characteristics which are essential for efficient nonlinear wave interactions.

One of the most common types of optical microresonators is a spherical silica glass microresonator, although its use is rather limited due to a relatively small nonlinear refractive index of the material. By adjusting the radius of a microsphere it is possible to shift the zero dispersion wavelength only in the long-wave direction, which poses difficulties in achieving dispersion curves needed for applications. However, adding a layer of another glass with higher linear and nonlinear refractive indices could significantly increase nonlinear coefficients and open up new possibilities for nonlinear effects. As the thickness of the layer becomes an additional parameter, the dispersion can be controlled.

In this study we analyze possible degenerate four-wave mixing (FWM) effects in silica (SiO_2) glass microspheres with a thin ($\sim 1-3$ µm) (1-X)·SiO_2-X·GeO_2 germanosilicate coating, where X is the relative molar concentration of germanium dioxide. To analyze possible generation wavelengths we used the phase-matching condition derived from the Lugiato-Lefever equation for electric field in a cavity with given dispersion and nonlinear coefficients [2]. As the microresonator dispersion curves varied significantly for different parameters, higher dispersion orders were used in the phase-matching condition. It is important to note that non-degenerate FWM processes can also be analyzed using this method.

For the pump wavelengths $\lambda_p = 1.28-1.68$ µm, we found multiple microresonator configurations allowing to meet the phase-matching conditions in a wide range, often limited only by the long-wave transparency of the glass of approximately ~ 2.7 µm. Additionally, it was also possible to find adequate microresonator parameters to achieve broad tunability ranges for pump wavelengths in common telecom bands as, for example, in SCL-band (1.460–1.625 µm). The obtained theoretical results for simple silica microspheres without coating were verified experimentally [3]. In comparison with a simple silica microsphere, the proposed microresonator composition potentially allows broadening of the generation range and finer parameter tuning to achieve a target wavelength as a result of the four wave-mixing process.

This work is supported in part by the RSF, Grant № 20-72-10188 and in part by the Megagrant of the Ministry of Science and Higher Education of the RF, Contract № 14.W03.31.0032.

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Unique dual-pulse diode laser with controllable (3-20 ns) interval & duration for the eye-safe lidar

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It is known that the new-generation compact lidar based on a pulsed diode laser and a time-gated SPAD (single photon avalanche photodiode) detector has opened wide prospects for environment sensing by eye-safe (< 1 µJ/cm^2) microjoule pulses [1]. It is clear that sensitivity of the lidar could be increased 10–20-fold by applying a time-gated SPAD detector like in Raman spectroscopy [2]. The SPAD detector allows for precise temporal synchronization for the pump-probe technique by dual-pulse fluorescence excitation [3].

The diode laser pulse duration can be shortened below 1 ns by decreasing the discharge capacity and by using a fast avalanche transistor key. We have developed a separate pump generator (Fig.1a) creating short pulses of current (2-20 ns) for an AlGaAs laser diode (SPL PL90_3, OSRAM). The oscillograms (Fig.1b) show that the dual-pulse generation and pulse shortening from 20 to 3 ns was achieved (Fig.1b) when decreasing C4 capacity from 1532 to 82 pF. The minimum pulse length (∼3 ns) with ∼ 0.2 µJ energy was achieved with time delay around ∼5 ns when 82 pF capacity was used. It should be noted that the pulse rise time remains small (∼1–2 ns) and the first-to-second pulse amplitude ratio is ∼3:1 for dual-pulse generation mode (Fig.1c) which is good for the pump-probe technique.

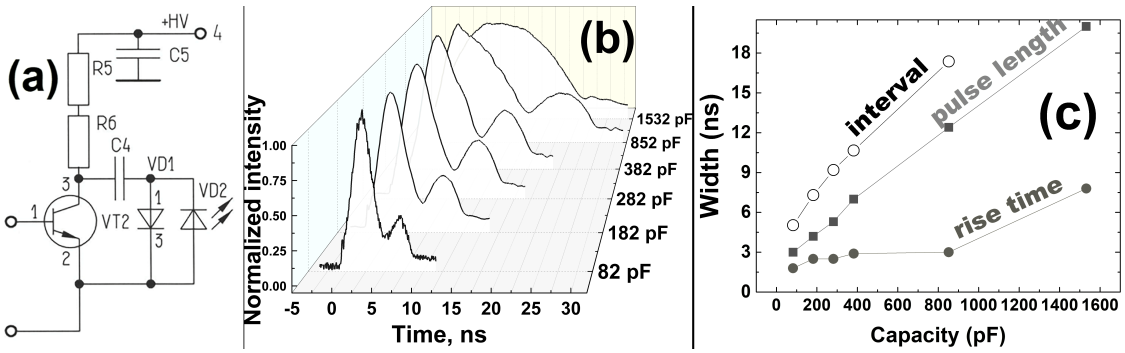

Figure 1: Current pulse generator circuit diagram (a); laser pulse shortening (b); laser pulse parameters dependence on discharge capacity C4: rise time (filled circles), pulse length (filled squares) and interval between the first and second pulses (open circles).

The authors gratefully acknowledge the financial support of the Russian Science Foundation (project № 19-19-00712).

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Low coherence lasing in dense mixtures of solid-state microparticles and liquids

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Studies of lasing in dense mixtures of microparticles and immersion liquids (slurry lasers) have been started in works [1,2]. Near 1 mJ, 20 ns pulses of stimulated emission were obtained both in slurries on LiF particles with F_2 color centers and on laser dyes solutions with transparent LiF particles. A characteristic feature of such lasers is the reduced spatial coherence of radiation in a directional laser beam. Such radiation is in demand for different applications: speckle-free laser imaging, focusing through scattering media, etc. After amplification it can be used in experiments on laser-matter interactions and in material processing. We present results of experiments with slurry lasers on PM567, Rh101 and DCM dyes in isobutanol with LiF grains. The state of immersion in slurry can be controlled by adding small amounts of another solvent or by temperature change. It was found that the degree of spatial coherence γ, measured with a double-slit Young interferometer, depends on the state of immersion. With a good immersion, γ is above 0.1 but significantly lower than γ for a dye laser without grains. The radiation spectrum is structured and the output beam profile has a core and several concentric rings with intensity maxima corresponding to the transmission maxima of the Fabry-Perot interferometer formed by a laser cavity. In this case up to 80% of the low-coherence radiation energy is contained in the central region of the laser beam. When detuning from the best immersion, γ decreases to 0.02 (measurement limit), while the lasing spectrum becomes smooth and beam divergence increases several times, remaining within 80 mrad, Fig. 1. Study of the space-time structure of the slurry laser radiation is in progress.

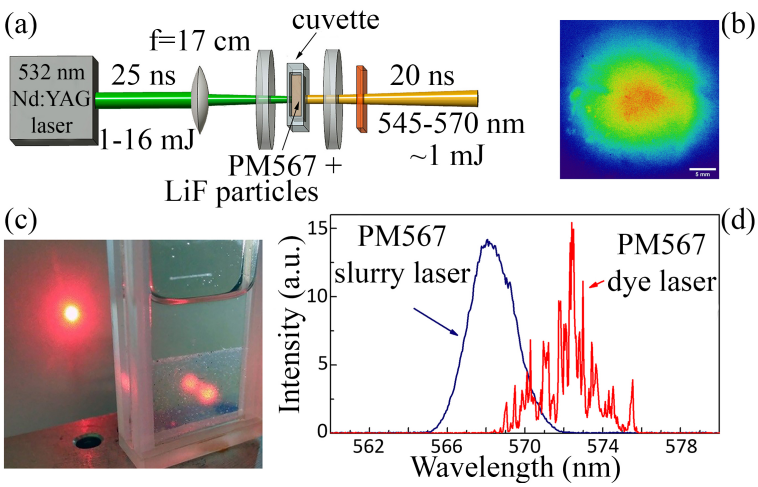

Figure 1: a) Slurry laser scheme; b) Beam profile in the focus of a lens with f = 23.5 cm; c) Cuvette with slurry; d) Emission spectra of the PM567 slurry and PM567 dye lasers.

This work was supported by the Russian Foundation for Basic Research (Grant No. 19-02-00344).

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Tunable dual wavelength and narrow linewidth laser using a single solid-state gain medium in a double littman resonator

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The laser emission characteristics of Littman-grating ultraviolet Ce: LiCAF crystal lasers pumped by the fourth harmonics of a Nd:YAG Q-switched laser at 266 nm also have studied by solving the multi-wavelength rate equations. The obtained results show that using a Littman resonator configuration, very narrow linewidth UV laser emission (a few picometers) can be continuously tuned over a wide spectrum from 278 to 315 nm. Furthermore, independently tunable two-wavelength UV laser emission is also achieved by a double-Littman resonator laser configuration using the same Ce:LiCAF crystal and pumping optics. This wavelength region has important applications in monitoring the concentration of sulphur dioxide and ozone in the atmosphere using differential absorption lidar which require two closely spaced wavelengths with linewidths of 100 pm or less. The proposed scheme can be extended to other wavelength regions using an appropriate laser gain medium, enabling other applications requiring a tunable dual wavelength and narrow linewidth laser.

Free space strange and unipolar waves in electrodynamics and acoustics

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Generation of unipolar electromagnetic pulses and simulation of their propagation and interaction with matter is demanded by diverse research and application fields. They are protection of electronic circuits and computers, physics of lightning, prediction of earthquakes, fundamental atomic physics on the timescale 10-14 - 10-16 sec, time resolved spectroscopy of chemical processes, coherent control of elementary particles and quantum systems and others [1-7]. Lasers since the late 90ies became a well accessible instrument to obtain few cycles, down to one pulses, i.e. to path the way to unipolar electromagnetic waves [8]. The terms unipolar (single sign) and strange waves were first introduced by E.G. Bessonov [9] even before the era of ultrafast optics. He studied the far zone of radiation field produced by electrons in accelerators and by various elementary processes in vacuum and media. This paper analyzes the conditions for strange and unipolar pulses in the free space.

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Powerful diode side-pumped Er:YLF laser operation regimes

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There is a growing demand for powerful mid-infrared laser sources. In particular, 3 µm nanosecond lasers are a convenient tool for optical pumping of optical parametric oscillators, femtosecond Fe:ZnSe-based amplifiers[1], as well as picosecond CO_2 amplifiers. The unique feature of 3 µm radiation is also its extremely high absorption in water and biological tissues (absorption coefficient α ~104 cm^{-1}). This fact gives rise to a large number of applications of such sources in technological, scientific, and medical fields.

Despite the rather long history of the development of solid-state 3 µm lasers, some lasing regimes are still challenging for some fundamental and instrumental reasons. Some applications require high peak power with high pulse energy, while others prefer high average power at repetition rates of tens to hundreds of Hertz. High pumping of heavily-doped crystals and a large quantum defect leads to a high thermal load on the active element, the appearance of thermo-optical distortions that distort the beam mode and lead to a breakdown of the optics [2]. The self-termination of the 3 µm laser transition also limits their application for Q-switching. In addition, high damage threshold electro-optical and acousto-optical materials with low losses in the mid-IR range remain almost inaccessible [3]. Thus, new approaches in laser design and the search for new active media are required.

The active medium Er:YLF, in comparison with other erbium-doped crystals, has many favorable properties for high average and peak power pulsed generation. The long lifetime and natural anisotropy of the crystal are favorable for storing energy and converting it into the Q-switched pulse. A weak negative thermal lens provides greater control over the beam caustics.

We report on the study of the operation of a DPSS Er:YLF laser in the regimes of free running and electro-optical Q-switching based on the KTP crystal. In Er:YLF, the processes of up-conversion and cross-relaxation have a strong influence on the generation dynamics. The presence of several transitions between electronic sublevels provides multi-frequency generation in the range of 2.67–2.85 µm. Such mid-IR nanosecond sources are promising for many fundamental and industrial applications including laser-induced backside wet etching (LIBWE), 3D bioprinting by laser-induced forward transfer (LIFT), and the study of the extreme state of water.

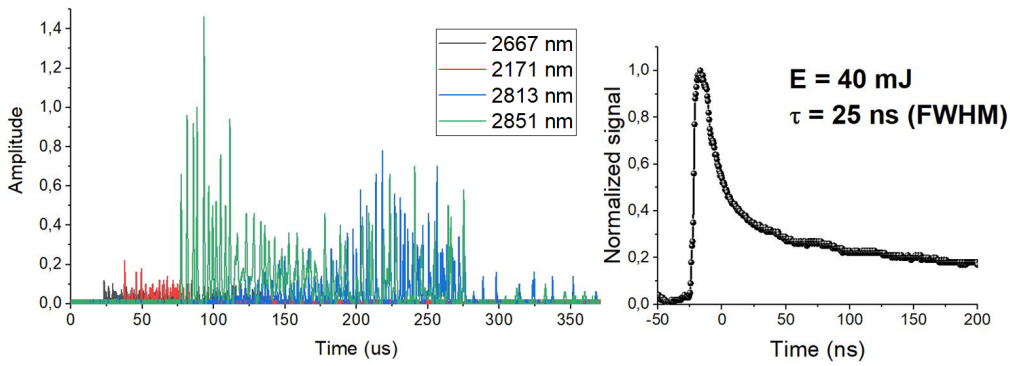

Figure 1: Spectral composition of free-running generation (left) and Q-switched pulse temporal shape of Er:YLF laser (right).

Er:YLF DPSS laser development is supported by Russian Foundation for Basic Research (RFBR) (№ 18-29-20074). The study of the 3 µm laser pulses interaction with water is supported by Russian Scientific Foundation (RSF) (№ 17-72-20130).

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Efficiency of excitation of a quantum two-level system by free space ultrashort laser pulse

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The problem of formation of few and subcycle laser beams for controlled action of radiation on matter is considered. The presence of the spatio-temporal couplings (SPC) makes it necessary to turn to models based on exact solutions of Maxwell's equations.

In this work, we use one of these solutions, which correspond to a collapsing and then expanding electromagnetic pulse of finite energy. As it propagates from the far zone to the near zone, the pulse shape and spectrum change in accordance with the SPC. Taking into account this effect, the maximum achievable fields and the efficiency of energy transfer from quasi-monochromatic and ultrashort EM pulses to a quantum two-level system are found in terms of the spectral parameters of the incident beam.

It follows from the results that the measurement of the transition probability in a two-level system under the action of a pulse beam can be used to diagnose the pulse itself, in particular, to establish the presence of unipolarity.

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Applicability of second order nonlinear crystals for broadband ultrashort mid-IR pulse generation

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Development of laser sources emitting broadband ultrashort pulses in the mid-IR is of great interest for fundamental research and applied purposes such as gas analysis, sensing of the atmosphere, laser chemistry and others. Generation and amplification of femtosecond pulses with octave spectrum can be produced in a second order nonlinear crystal [1, 2]. This technique was experimentally demonstrated with $ZnGeP_2$ and GaSe crystals [2]. In this work we numerically investigated applicability of eleven nonlinear crystals for broadband ultrashort mid-IR pulse generation and compare their efficiencies.

We considered following nonlinear crystals: $AgGaS_2$, $AgGaSe_2$, $BaGa_2GeSe_6$, $CdGeAs_2$, $CdSiP_2$, GaSe, $HgGa_2S_4$, $PbIn_6Te_{10}$, and $ZnGeP_2$. The crystal properties were calculated: phase-matching angles, effective nonlinearity, spectral and angular phase-matching acceptance, and group velocity mismatching. Conditions of wide spectral acceptance required for broadband mid-IR emission generation are found for each crystal, as well corresponding spectral bandwidths of idler and signal wave generations. Efficiencies of the crystals for broadband signal and idler wave generation are estimated with integral figure of merit, which was previously verified with broadband sum frequency generation of multiline CO-laser [3].

It was found that for broadband generation within wavelength range of about 5.4—6.5 µm the $ZnGeP_2$ is the most suitable crystal. The $HgGa_2S_4$ is attractive for a shorter wavelength range 3.5–5.1 µm. The $CdGeAs_2$ and $PbIn_6Te_{10}$ crystals can provide generation, respectively, within the widest spectral range at the longest wavelengths about 6.5–11 µm.

The research was supported by the Ministry of Science and Higher Education of the Russian Federation № 121031300155-8.

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Multifrequency stimulated Raman scattering in ethanol and glycerol under picosecond excitation

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Ethanol and glycerol are among the simplest alcohols that are actively used in nonlinear optics [1, 2]. Due to the high radiation resistance and relatively high Raman shift of the highest Q mode of spontaneous Raman scattering, these media are of great interest for the generation of frequency combs in a wide spectral range under stimulated Raman scattering (SRS). To excite multifrequency SRS, we used the main, second, and third optical harmonics of a repetitively pulsed YAG:Nd^{3+} laser generating ultrashort pulses at wavelengths λ = 1064 nm (80 ps), λ = 532 nm (60 ps), λ = 355 nm (55 ps), respectively. Upon excitation of SRS in ethanol by radiation with a wavelength of λ = 532 nm, two Stokes and one anti-Stokes satellites with frequency shifts of 2950 cm^{-1} were recorded. In glycerol, using a green laser line, it was possible to register two Stokes SRS satellites with the same Raman frequency of 2950 cm^{-1}. The use of the third harmonic of the laser at λ = 355 nm made it possible to register two equidistant Stokes components of SRS, as well as their overtones.

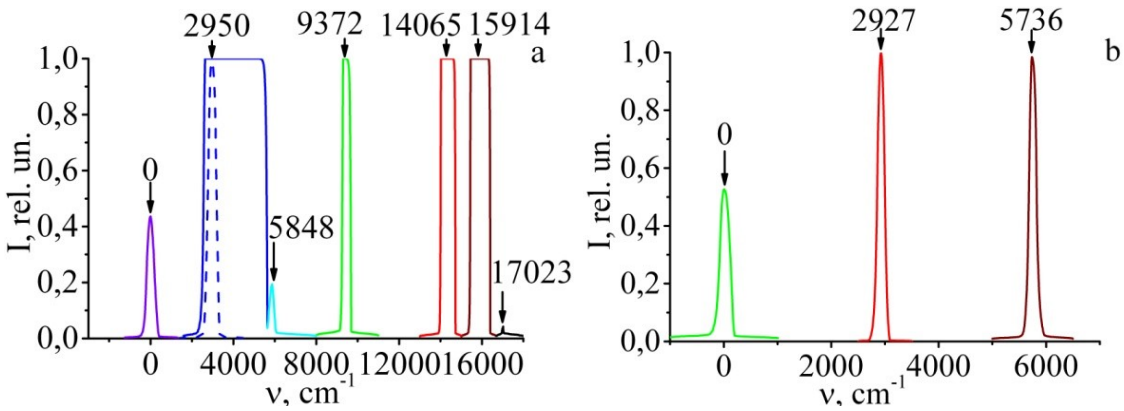

Figure 1: Normalized spectra of multifrequency SRS in ethanol (a) and glycerol (b) upon excitation by laser radiation with a wavelength of λ = 355 nm and λ = 532 nm, respectively.

The reported study was funded by RFBR and BRFBR (projects № 20-52-00002 and № 20-52-04001).

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Laser pulse shortening via cascade stimulated Raman scattering in crystals on combined vibrational modes under picosecond synchronous pumping

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Stimulated Raman scattering (SRS) in crystals is a simple and efficient method of nonlinear optical conversion of laser radiation, which doesn't require phase matching to be maintained. However, using ultrashort laser pulses with duration shorter than dephasing time of vibrational modes in crystals (∼1-10 ps) is a problem for efficient Raman conversion because of falling Raman gain in a transient regime of SRS. Using synchronous pumping technique for crystalline Raman lasers allowed overcoming the problem of low transient Raman gain and obtaining efficient generation of high-repetition-rate subpicosecond Stokes SRS pulses under femtosecond laser pumping [1]. Recently such lasers were efficiently applied for multicolor two-photon 3D imaging in living tissues [2].

We present another method [3] of strong laser pulse shortening down to subpicosecond duration under picosecond synchronous pumping via cascade Raman conversion with combined frequency shift on several vibrational modes in crystals. The method allows using lower cost and simpler picosecond solid-state lasers than femtosecond lasers for pumping to obtain high-repetition-rate multicolor radiation pulses with subpicosecond duration. Scheelite, zircon, and whitlockite families of crystals were comparatively studied [3, 4] as the active Raman media having two intense Raman modes of stretching and bending internal symmetric vibrations of anionic groups. Mechanism of laser pulse shortening is cascade-like. The first cascade is extracavity Raman conversion on the most intense stretching Raman mode of an active crystal into a first Stokes SRS component. The second cascade is intracavity Raman conversion of the first Stokes component into a second Stokes component with combined (stretching + bending) frequency shift and strong pulse shortening down to dephasing time of the widest bending Raman mode. We found series of crystals with bending line widths wider than 10 cm^{-1} such us YVO_4 (10.4 cm^{-1}), $SrMoO_4$ (10.5 cm^{-1}), $GdVO_4$ (24 cm^{-1}), $Ca(VO_4)_2$ (50 cm^{-1}) for laser pulse shortening shorter than 1 ps (inverse linewidth). For the Raman-active $GdVO_4$ crystal, a first result of subpicosecond SRS generation (860 fs at 1228 nm) under synchronous pumping by 36 ps, 1063 nm Nd:$GdVO_4$ laser has been achieved [3]. It was found that pulse shortening happens down to inverse width of homogeneously broadened part of the Raman line. Our latest study is to search and use crystalline solid solutions with different cations (Ca + Sr) or anions (MoO_4 + WO_4) to widen of the bending Raman mode while maintaining its one-mode behavior that reduces the subpicosecond SRS pulse duration.

The research was supported by the Russian Foundation for Basic Research – Project № 19-02-00723.

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Ultrashort pulsed mid-IR lasers for science and technology

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The talk reviews recent progress in ultrashort pulse generation in the mid-IR wavelength range between 2 µm and 4 µm based on fiber and solid-state lasers and amplifiers, highlighting the most recent achievements in the Laser Physics Group at NTNU such as a high pulse energy (up to 60 nJ) Cr:ZnS oscillator, mJ-class level femtosecond/picosecond Ho-fiber MOPA, and Yb- and Tm-fiber MOPA based OPGaP frequency combs. The shortest pulses that we could recently generate around 4 microns reach only three optical cycles in duration at the highest reported output power of 250 mW and 50 % efficiency.

The built-in tunability of femtosecond pulses in Tm-fiber laser, mJ-class pulse energies from the Ho-fiber based MOPA, high quality frequency combs as well as the ability to produce supercontinua directly from the laser are making this novel mid-IR laser technology particularly attractive for industrial applications demanding either high quality and fine material processing or ultrahigh sensitivity measurements. The application areas include, but are not limited to microelectronics, photovoltaics, THz generation, confocal nonlinear microscopy and neurosurgery, as well as environmental, oil and gas sensing. In this talk we will discuss a few most interesting applications that benefit from the ultrashort pulsed mid-IR femtosecond laser sources, focusing on fine material processing of semiconductors in general and silicon in particular, as primary future application of ultrafast lasers operating above 2 microns.

Towards efficient mid-IR optical parametric amplifier pumped by Ti:sapphire laser

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Few cycle terahertz (THz) pulses of high field strength have attracted large attention due to the development of nonlinear terahertz photonics, which opens up new opportunities for exploring electronic subsystem dynamics far from equilibrium, ultrafast magnetisation, imaging and others. Efficient generation of intense THz radiation with field strength up to units of GV/m under optical rectification of the optical pump has appeared only recently with advent of organic crystals (such as DAST, DSTMS and OH1) [1]. However, dispersion properties of these materials require near-IR (1.2–1.5 µm) pump. Moreover, exceptionally high optical-to-THz generation efficiency approaching 6% was observed in DAST in the case of a 1.95 µm pump pulse due to low linear and multi-photon absorption in combination with cascaded optical rectification [2]. Unfortunately, today there are only a limited number of laser media capable of providing high-power (tens of mJ) femtosecond (less than 100 fs) pump radiation in the near and mid-IR wavelengths [3], [4]. Therefore, we consider the creation of the pump source for THz generation that can be obtained by wavelength conversion into infrared spectral region, based on terawatt Ti:sapphire laser system, which can provide a good scalability of the output energy. Recently, due to the availability of high-power laser systems, a "dual chirped optical parametric amplification" (DC-OPA) scheme has been proposed to create efficient terawatt parametric amplifiers [5]. Chirping of both pump and signal pulses allows to avoid unwanted cross- and self-phase modulation and overcome energy limitations associated with the available aperture size of BBO crystals. In this paper, we perform a theoretical comparison of direct OPA and DC-OPA schemes for wavelength conversion into the near-IR region (1.3–2.2 µm).

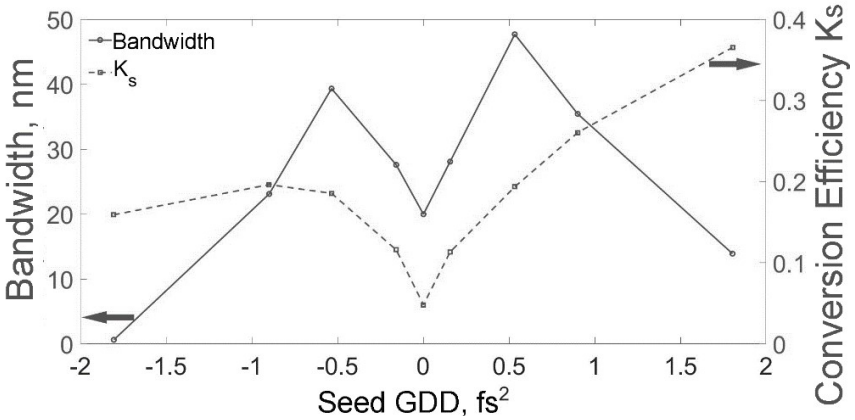

Figure 1: Signal pulse conversion efficiency (dashed line) and bandwidth (solid line) obtained from the DC-OPA for different values of the seed GDD (group delay dispersion).

To quantitatively evaluate the conversion efficiency and the bandwidth of the OPA and DC-OPA schemes, we carried out numerical calculations based on coupled wave equations describing three-wave interactions. The simulation was performed for 800 nm, 50 fs, 60 mJ pump radiation and considered type II BBO crystal as the most promising one. We have found that in the case of direct OPA scheme, the signal (1.3 µm) and idler (2 µm) pulses can be produced with pulse durations of 34 and 32 fs, respectively, which contain less than 5 optical cycles, with a total conversion efficiency of 40%. In the case of DC-OPA scheme the

total conversion efficiency increases up to 60%, but a decrease in the spectral bandwidth of the obtained pulses is observed (Figure 1). An increase of the spectral bandwidth can only be achieved with a decrease in conversion efficiency. Therefore, in the case of a 100 mJ class Ti:sapphire laser with a pulse duration of 50 fs, direct OPA scheme based on type II BBO crystal is the most promising approach due to its high efficiency and the ability to generate few-optical-cycle pulses. In three stage OPA configuration one may expect up to 6 mJ and 10 mJ in idler and signal waves, respectively. This OPA scheme is now under commissioning and we are working on optimization of the first two stages.

This work has been supported by Russian Science Foundation Project № 20-19-00148.

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Towards the analysis of attosecond dynamics in complex systems

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The progress in laser technology over the last decades has opened up new avenues for the exploration of properties of clusters and molecules. A laser pulse is characterized by its frequency but also by the laser intensity as well as the laser time profile. The latter can now be tailored up to time scales of the order of magnitude of electronic motion and even below. This allows the follow up of the detail of electronic dynamics at its own "natural" time. We shall focus in this presentation on the recent explorations of electron dynamics down to the attosecond time scale. Some experimental cases can be well reproduced by time dependent microscopic theories. For accessing a detailed explanation of observed trends we introduce a schematic model which surprisingly enough provides a remarkable account of experimental trends. It shows in particular that the response of the system is heavily biased by properties of the laser used for exciting and testing the system, especially its IR component. Using the ideas developed in the schematic model we can reanalyze former computed data and show how much one can attain system's properties.

We then focus on a specific pump and probe setup. By recording observables of electron emission we analyze the response of small metal clusters and organic molecules to a pump probe setup using an IR fs laser pulse as pump followed by an attosecond XUV pulse as probe. As observables, we consider total ionization, average kinetic energy from Photo Electron Spectra (PES) and anisotropy parameters from Photo-electron Angular Distributions (PAD). We show that these signals can provide a map of the system's dynamical properties. The connection is especially simple for metal clusters in which the response is dominated by the Mie surface plasmon. The case of organic molecules is more involved due to the considerable spectral fragmentation of the underlying dipole response. But at least the dipole anisotropy from PAD provides a clean and robust signal which can be directly associated to system's properties even reproducing non-linear effects such as the change of spectra with excitation strength [6].

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Optically transparent coatings based on refractory compounds for protection of laser-induced damage

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The development of high-power and high-energy laser technologies has led to an increase in work on the development of thin optical films that are resistant to the intense action of a high-power laser [1]. An important parameter for such films is the laser-induced damage threshold (LIDT) [2]. The LIDT of thin film is influenced by material composition, deposition method, defect structure, and surface contamination. The method of magnetron sputtering with preliminary ionic cleaning will make it possible to obtain defect-free films with high adhesion and the absence of contamination. This work is devoted to the obtaining of thin films based on refractory metal compounds (Zr, Mo, Hf, etc.), the study of their optical characteristics, and resistance to the laser action of different power.

Nanocomposite ceramic films were deposited by magnetron sputtering by direct (DCMS) and pulsed current (PMS), as well as high-power impulse magnetron sputtering (HIPIMS) in Ar + N_2 gas mixtures. The structure, chemical, and phase composition of the thin films were investigated by scanning electron microscopy, energy dispersive spectroscopy, X-ray diffraction, transmission electron microscopy and glow discharge optical emission spectroscopy. The mechanical characteristics of films were studied by the method of nanoindentation. The adhesive strength was determined by scratch testing. Optical properties such as transmittance, refractive index and reflectance were measured with the Agilent + UMA Cary 5000 instrument over the wavelength range 200 to 2500 nm. The study of resistance to laser action was carried out at different laser power. For research at low laser power, the device for Raman spectroscopy Ntegra Spectra NT-MDT, equipped with a red laser with a wavelength of 633 nm and power in the range of 10–24 mW, was tested for the first time for laser resistance. Studies were also carried out on Trotec Speedy 400 flex and LaserPRO Spirit devices with a maximum power of 100 W.

The results showed that the thin films with a high nitrogen content showed an amorphous structure, consisting mainly of BN, with fine-grained inclusions of metallic phases. Due to film thickness, the concentration of the amorphous phase, and chemical composition, higher optical characteristics are achieved. For films high values of the transmittance were observed in the range of 80–100%. As a result, thin films based on refractory metal compounds (Zr, Mo, Hf, etc.) showed good resistance to the laser-induced damage.

The work was supported by the Russian Foundation for Basic Research (Agreement № 19-08-00187).

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Mid-infrared fiber laser pumped Fe:ZnSe lasers

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High power lasers with the wavelength range of 2.5–5 µm have many potential applications, for example, in laser processing for materials which have low absorption at the near-infrared and the visible wavelength range, and in dentistry and surgery because the absorption coefficient of biological tissue containing water is very high at the mid-infrared region.

Here, we review mid-infrared Er:ZBLAN fiber lasers [1–4] and Fe:ZnSe lasers pumped by fiber lasers [5–7]. We developed high-power Er:ZBLAN fiber lasers whose output power exceeds 30 W at 2.8 µm and applied 10-W-level fiber lasers for pumping Fe:ZnSe lasers that cover a wavelength range from 4 to 5 µm. We demonstrated that ZnSe crystals reach their highest potential with a high-beam-quality fiber laser. This laser technology paves the way for the development of powerful mid-IR sources for cutting-edge fields in industry, medicine, and science.

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Intracavity Stimulated low-frequency Raman scattering — a spectroscopy method in the gigahertz frequency range

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Low-frequency Raman scattering (LFRS) is the spontaneous inelastic scattering of light caused by the natural (localized) acoustic vibrations of nano- and submicron particles [1, 2]. It is a method of obtaining information about the shape of nanoparticles, their characteristics of interaction with electromagnetic radiation, and the size distribution.

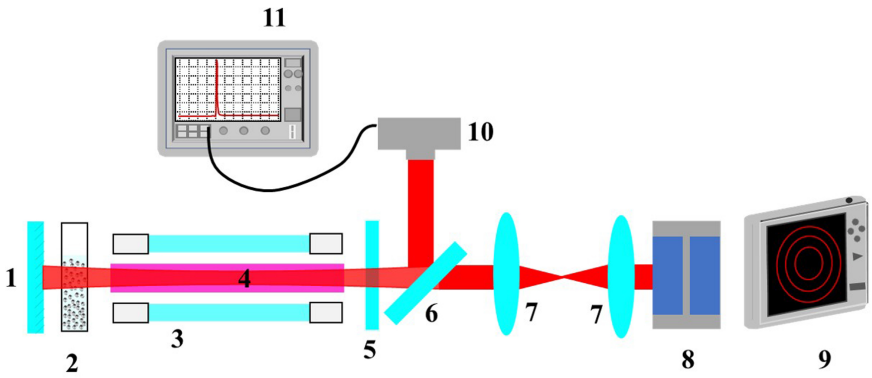

Figure 1: The experimental setup for excitation and registration of intracavity SLFRS: 1 – opaque mirror; 2 – a cell filled with a suspension of monodispersed polystyrene nanoparticles in water; 3 – lamp; 4 – ruby crystal; 5 – output mirror; 6– glass plate; 7 – lenses; 8 – Fabry-Perot interferometer; 9 – camera; 10 – photodiode; 11 – oscilloscope.

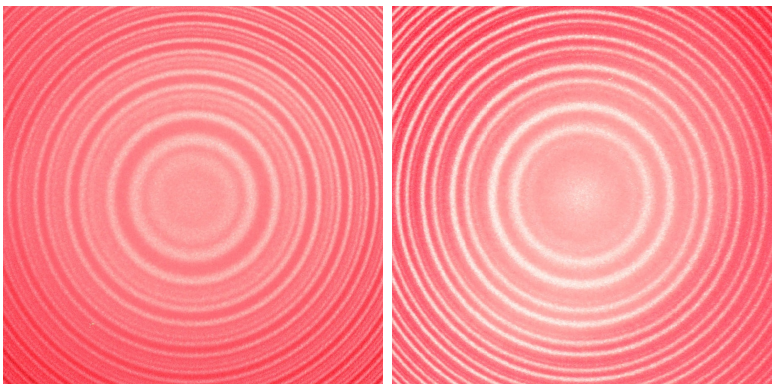

Figure 2: Fabry-Perot interferograms of free generation (a) and intracavity SLFRS of polystyrene in water (b)

Stimulated low-frequency Raman scattering (SLFRS) is a stimulated process of LFRS with a large conversion efficiency of up to 70 % and a narrow spectrum. This work proposes a new intracavity scheme for SLFRS by analogy with intracavity Raman scattering, one of the most sensitive methods for diagnostics of various kinds of substances today (see. fig. 1). An experimental study of intracavity SLFRS in aqueous suspensions of monodispersed polystyrene nanoparticles of various sizes (100–500 nm) was presented. A nanosecond ruby laser with a wavelength of 694.3 nm and a narrow spectral line of 0.015 cm^{-1} was used as

excitation radiation. The energy of the laser pulse, its duration, and spectral composition were monitored for all measurements. Fig. 2 shows typical Fabry-Perot interferograms of free generation of a ruby laser (a) and intracavity SLFRS of polystyrene in water (b). Spectral lines with natural acoustic frequencies in the gigahertz range corresponding to polystyrene particles eigenvibrations appeared in the laser spectrum when the cell with nanoparticles suspension was placed in the laser resonator.

The experimentally obtained frequency shifts of the Stokes intracavity SLFRS components correspond to those calculated values which were obtained using the Lamb approach, taking into account the influence of the environment.

It is experimentally shown that intracavity SLFRS has a much higher potential, allowing one to obtain components of a higher order than in conventional SLFRS.

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Time-domain acousto-optic modulation and direct characterization of chirped laser pulses

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Spectral shaping of phase-stretched broadband laser pulses is an effective way to obtain fast and programmable modulation in time domain. Acousto-optic spectral modulation of chirped laser pulses enables arbitrary optical waveform generation with multi-GHz bandwidth [1]. In this work, we report direct time-domain assessment of a programmable millijoule-level pulse source based on a high-resolution pulse shaper and a Ti:sapphire regenerative amplifier. Laser pulse envelopes were synthesized at the stretcher input with a high-resolution acousto-optic dispersion delay line (AODDL) designed and fabricated in-house. The chirped pulse envelope was measured directly at the amplifier output without compression using a picosecond streak camera. Binary spectral phase modulation was applied to assess the resolution of the pulse shaper and measurement instrumentation. The minimum resolvable element in the streak camera trace was found to be 1.5 ps.

Arbitrary modulation with multiple stopbands in the spectrum was studied in a series of experiments. Binary spectral modulation was used to assess pulse fronts in transient response measurement mode. AODDL transmission is a programmable complex-valued function, which is implemented by specific phase-and-amplitude modulation of ultrasonic waveforms. Two algorithms for waveform synthesis were compared: explicit dispersive Fourier synthesis (DFS) algorithm and iterative Gerchberg-Saxton algorithm [2]. In both cases, modulation rise/fall time was found to be dependent of the sign and value of the second-order dispersion (SOD) of the ultrasonic waveform in the AODDL. The maximum SOD value was limited by the uncertainty relation for chirped pulses [3]. Average measured pulse fronts ranged from 3.6 to 6.7 ps depending on experimental settings. The shortest pulse fronts of 3.6 ± 0.6 ps were obtained using the DFS method for the pulses with 6.2 ps/nm linear chirp when the AO-induced SOD was negative.

The research was supported by the Russian Foundation for Basic Research (Project 18-29-20019).

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Section 7: Ultrafast laser technologies and structured light in micro-optics and nanophotonics

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Scope

Ultrafast nanophotonics in dielectric nano/microstructures

Ultrafast structured light

Femtosecond/picosecond laser writing in dielectrics

Novel optical materials for ultrafast photonics

Design and manufacturing of planar optical waveguide by femtosecond laser recording technique

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When developing planar waveguides (especially those with complex cross-section geometry), for example for integral photonics problems [1], it is necessary not only to choose the waveguide material, but also to determine the optimal method of its manufacture. On the one hand, the method should provide a pre-designed waveguide structure with a minimum error, on the other-it should be the least time-consuming and not greatly increase the cost of the final product. At the same time, laser pulses of various durations (femtosecond, subpicosecond, and short picosecond) are now used to solve the most advanced fundamental and applied problems of science, technology [2-4] and technology, including those related to modifying the surface and volume of various dielectrics to create functional nano - and micro-sized layers [5].

This work is devoted to the creation of a planar waveguide. Relying on the experience in the work [6], where is given a general treatment of refraction phenomena (anomalous refractive behavior, negative refraction e.t) in a transparent slab, which show that these interference effects impact the beam position on the rear face for any slab thickness, we can see an approximate formulas for low divergence light beams. They show the presence of anomalous refraction phenomena at any slab thickness, including negative refraction and flat lensing effects, induced by reflection at the rear face.

The combination of the physical and chemical properties of sapphire, quartz glass and diamond with the capabilities of femtosecond laser processing makes these materials attractive for the manufacture of any planar waveguides based on it, which are applicable for a wide range of applications, including those suitable for use in aggressive environments.

This work was supported by the Russian Science Foundation (Grant No. 21-79-30063).

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Lithium niobate nanophotonics

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Since the concept of photonic integration was created in 1969 [1], it is now clear that photonic integrated circuits (PIC) is becoming the ideal platform for numerous applications from microwave photonics, free space communication to optical networking and quantum computing. To make the dream of photonic integration come true, great efforts have been devoted to achieving a sustainable scalability similar to that of its electronic counterpart, as well as low propagation loss, high tuning efficiency and speed, and small bend radii. However, most material systems can hardly support all these requirements. Here, we report on fabrication of high quality (Q) factor microresonators on lithium niobate on insulator (LNOI) using photolithography assisted chemo-mechanical etching (PLACE), which holds the potential to realize high performance PICs of footprints merely limited by the wafer size. Optical microresonators with Q factors well above 10^8 and single mode optical waveguides with a propagation loss below ~ 0.03 dB/cm is fabricated on LNOI via PLACE. Highly efficient nonlinear processes from second harmonic generation (SHG), third harmonic generation (THG), optical parametric oscillation (OPO), optomechanics to comb generation, meter-long optical delay line, and active waveguide lasers and amplifiers are demonstrated. [2-3]. We believe that by developing the PLACE technique, large scale active/passive photonic integration is now getting mature and will be achievable soon.

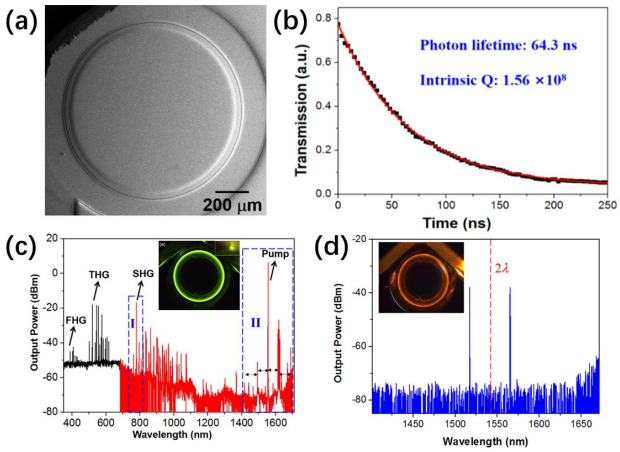

Figure 1: (a) The SEM image of the ultrahigh Q microdisk resonator on lithium niobate on insulator fabricated using PLACE. (b) The Q factor measurement.

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Resonance absorption in the bulk of dielectrics

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At high intensity, femtosecond laser pulses shaped as Bessel beams can generate void channels with extremely high aspect ratio inside dielectrics such as sapphire, glass or fused silica [1]. This can be usefully applied to glass cutting via the "stealth dicing" technique. This enables glass cutting a very high-speed. Many advances have been made in this regard by improving the shape of the Bessel beam to make it for instance elliptical, or to increase the aspect ratio of the beam so that stealth dicing now can be used to cut glass with thickness up to 1 cm [2]. However, the exact mechanism of energy deposition is hitherto largely unknown.

We report on several experimental campaigns where we measured single shot transmission and beam fluence sectioning, to evaluate how the energy is deposited inside dielectrics. We showed that extremely high absorption is reached during the laser pulse propagation (typically 50 percent) [3]. This raises the question why is so high energy density deposited while multi-photon absorption and avalanche ionization are generally relatively inefficient at femtosecond pulse durations.

We have conducted a series of laser plasma interaction modeling using a Particle-In-Cell (PIC) approach with EPOCH code [4]. We show that resonance absorption occurs in the bulk of dielectrics and that the transverse dimension of the laser-generated plasma is below the wavelength. This is assessed via comparisons between experimental and numerical simulation results over a number of different diagnostics. The resonance absorption phenomenon is an efficient collisionless mechanism to transfer energy from the laser wave to the plasma even for femtosecond pulse durations.

In conclusion, spatial shaping of laser pulses into Bessel pulses allows for accessing plasma profiles that are much steeper than those conventionally produced using Gaussian beams. With additional transmission measurements in a dual shot configuration, we could demonstrate that the high energy density deposited allows for transforming the material into warm dense matter in a temporal scale of 10 to 100 ps [3].

This research has received funding from H2020 European Research Council (ERC) under grant agreement 682032-PULSAR. This work was granted access to HPC resources PRACE (Project PULSARPIC PRA19_4980 and RA5614), TGCC (Projects A0070511001 and A0090511001), and Mésocentre de Calcul de Franche-Comté.

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Role of electronic interband transitions for the femtosecond laser energy deposition into dielectric materials

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Femtosecond laser pulses are an efficient tool for laser processing of dielectric materials including ablation and nano-frabication of various structures [1]. This processing is driven by the laser energy deposition into the material which induces phase transitions of matter [2]. To improve current techniques and design future experiments, an in-depth understanding of how the laser energy is absorbed is crucial.

Modeling efforts are mainly based on collisional absorption, i.e. the phonon- or ion-assisted absorption of photons by free electrons [3]. This absorption process corresponds in general to electronic intraband transitions. By using photoemission spectroscopy and modeling based on a kinetic Boltzmann approach, it has been shown recently that electronic interband transition should play a significant role on the laser induced electron dynamics, and thus on the laser absorption [4]. Despite first promising conclusions, this work has to be consolidated due to some assumptions including the simplified electron band structure which simply consists of one parabolic band.

The aim of this talk is to go further by properly describing the band structure and laser interaction through *ab initio* calculations [5]. For that purpose, the time dependent density functional theory is used to study the excitation dynamics of electrons in $\alpha-$quartz induced by two 10-femtosecond laser pulses exhibiting different wavelengths. The influence of the pulse-to-pulse delay and intensities couple on the electron excited density and their energy density is studied. The role of direct interband transitions for ionization (bridging bandgap) and excitation in the conduction band are highlighted. The efficiency of laser heating of conduction electrons is shown to be equivalent to collisional absorption within a 1 fs collision timescale, which is rather standard. These results thus definitely demonstrate the importance of interband transitions for the laser energy deposition into dielectric materials. The laser energy deposition into such dielectric materials can be controlled by adjusting the pulse-to-pulse delay and by taking advantage of the wavelength dependence of excitation efficiency.

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Optical properties of laser-induced plasma in water

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The plasma in water is a problem of great interest in solution chemistry, biology, and medicine [1,2]. Although the excess and then hydrated electrons are routinely produced via nonlinear absorption of intense light [3], our knowledge about its optical properties is rather limited. We know a lot about optical absorption of the aqueous plasma [4,5] and almost nothing about a real part of its optical polarizability. Here we present a study of refractive index of plasma in pure liquid water ionized by a strong optical field. Dynamics of water polarizability induced by a femtosecond laser pulse (duration 100 fs, wavelength 800 nm, intensity $\sim 10^{12}$ W/cm^2) was studied in the time window of ≈ 1.5 ns by means of a pump-probe interferometric technique which was developed for temporally and spatially resolved observation in solids.

Both the refractive index and absorbance were measured yielding quantitative data about the induced processes: solvation of excess electrons, geminate recombination and development of cavitation. The times of solvation and recombination are in good agreement with the data measured previously. The linear polarizability of the electron in the hydrated state was estimated to be ≈ 600 A^3.

The dependence of refractive index change and induced absorbance on pulse energy proved a five-photon absorption, in agreement with the well-known 6.5 eV threshold of optical ionization of pure water [6]. A strong saturation of plasma generation was revealed at $E \approx 0.7$ µJ. On the other hand we found that the number of electrons produced by NIR field in the cavitation regime ($< 10^{19}$ cm^{-3}) is much lower than was previously thought.

The detailed study of dynamics of plasma polarizability was performed also for 2-nd and 3-rd harmonics of the pump. It demonstrated a strong effect of wavelength on the cavitation intensity. This and other observations indicate that the dominant channel of energy deposition, which eventually results in the cavitation, is induced absorption, which is the linear absorption of the plasma of prehydrated electrons created by the pulse itself. Thus, the relatively low-dense plasma impels a significant increase in the temperature inside the affected microvolume.

Presented data provide an experimental basis for theoretical models that describe the interaction of intense femtosecond pulses and induced plasmas in aqueous media. This work was supported by the Russian Science Foundation (grant 19-12-00255).

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Transport-of-intensity equation in photonics problems

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Recently the transport-of-intensity equation (TIE) as a phase imaging method turned out as an effective microscopy method that does not require the use of high-resolution optical systems and a priori information about the object [1].

In practice, phase-contrast and differential interference-contrast microscopy methods are used to visualize weakly absorbing samples. However, these methods have one important drawback, which is that the measured intensity has a nonlinear and, therefore, non-convertible relationship with the sample phase. And without information about the phase, it is impossible to determine the morphology of the samples under study, such as size, optical thickness, strands, etc. These limitations, combined with the advent of digital image sensors and advances in information optics, have led to the emergence and expansion of the field of phase measurements and quantitative phase imaging techniques, which combines innovations in optics, imaging theory, and computational techniques to quantitatively display phase information. different designs. Despite the advances in optical interferometry, other methods of phase imaging have developed over the decades, for example, interferometric microscopy and digital holography, low-coherence holographic methods, or white light interferometric microscopy. Unlike the above methods, there is a category of methods for extracting phase from intensity measurements in a non-iterative manner. Compared to traditional interferometric methods, TIE has many advantages, no reference beam, simplicity of calculation, the ability to work with temporal/part-time coherent beams, no need for phase sweep and complex optical setups, and a stable measurement environment. The use of TIE turns out to be sufficient when the problem is posed of holographic reconstruction (albeit digital) of only three characteristics of the object field: amplitude, phase, and wavelength.

This work was supported by the Russian Science Foundation (Grant No. 21-79-30063).

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2D perovskite micro-optics and advanced microlasers enabled by direct femtosecond-laser printing

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Halide perovskite microcrystals, whiskers and plates, have appeared as novel building blocks for photonic integrated circuits. Particularly, smooth morphology, high quantum yield and bandgap tunability make them one of the most prospective candidates to develop tunable microscale optical elements and visible-range microlasers. Here, we show that femtosecond laser irradiation of single-crystal halide perovskite $CsPbBr_3$ allows for its precise and ultraclean ablation fully controlled at subwavelength scale by the intensity and polarization distribution of the complex laser field applied.

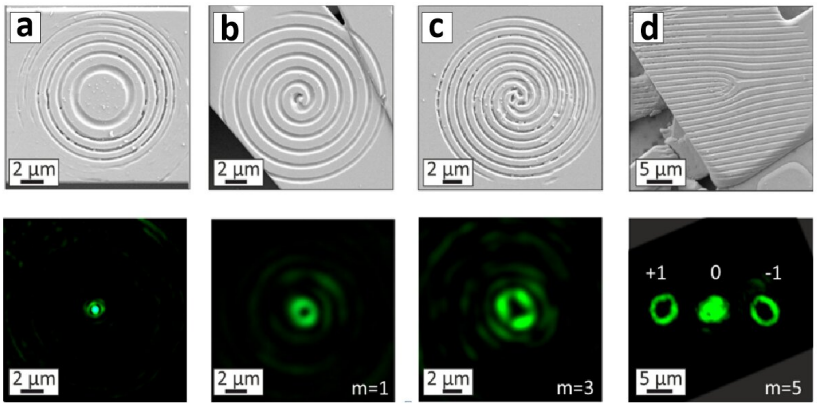

Figure 1: (a-d) Top-view SEM images of the $CsPbBr_3$ microcrystal surface imprinted with various micro-optical elements including Fresnel zone plate (a), spiral axicons (b,c) and binary fork-shape grating (d) of the as well as corresponding focal-plane intensity distributions at 515 nm excitation wavelength showing focusing performance of the imprinted 2D perovskite micro-optics.

Indeed, the extremely low thermal conductivity (over 300 times lower than that of silicon) and ultrafast thermalization rate makes it possible to reduce heat-affected zone and avoid melting layer contribution, while the high refractive index (larger than 2) provides high spatial resolution in case of irradiation of pre-patterned focusing perovskite nanostructures. These features allow for direct imprinting of the incident laser field at wavelength λ =515 nm, creating micro-lens and various light-emitting metasurfaces with deeply subwavelength spatial resolution (down to $\lambda/7$) [1]. Moreover, we demonstrated high-throughput fabrication of advanced binary microscale optical elements for nanofocusing as well as generation of high-order optical vortex beams.

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Ultrafast laser additive manufacturing: plant based organic and crystalline inorganic 3D nanostructures

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In this talk I will present recent updates on light-matter interaction mechanisms under spatio-temporally confined light and its applications targeting for 3D printing [1]. Fresh results uncovering photopolymerization dependence on irradiation wavelength will be reported [2]. Finally, realization of ultrafast laser additive manufacturing out of plant based resins [3], ceramics [4] and crystalline materials will be demonstrated [5].

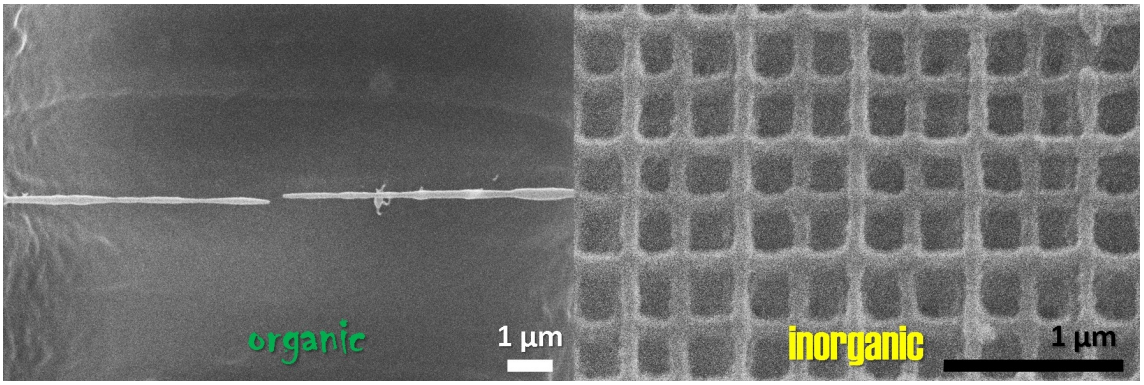

Figure 1: An examples of true 3D nano-structures produced out of organic and inorganic materials: a sub-100 nm suspended spike of vegetable oil-based thiol-ene/thiol-epoxy resin and a $\sim$ 65 nm linewidth lattice of Si/ZrO_2 crystalline phase substance, on left and right sides, respectively.

In Fig. 1 two benchmarking 3D nanostructures are shown out of pure organic and inorganic substances obtained via ultrafast laser direct writing lithography technique.

A financial support from the EU ERDF, through the INTERREG BSR Programme, (ECOLABNET project (#R077)) and EU Horizon 2020 research and innovation programme LaserLab Europe under grant agreement № 871124 is kindly acknowledged.

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New nonlinear photographic materials with fluorescent imaging

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These materials are used in scientific research [1, 2, 3] for photographing the spatial pattern of highly nonlinear interaction of intense laser radiation with transparent media, visualized after photographing in photoluminescent radiation. Such an application is relevant due to the fact that complex self-action processes occur during highly nonlinear interaction of optical radiation and matter, such as self-focusing, multiple filamentation of radiation, and others that radically change the initial spatial configuration of the radiation field. An experimental study of the spatial configurations of the field in matter realized in such interactions is of considerable interest for laser physics and nonlinear optics.

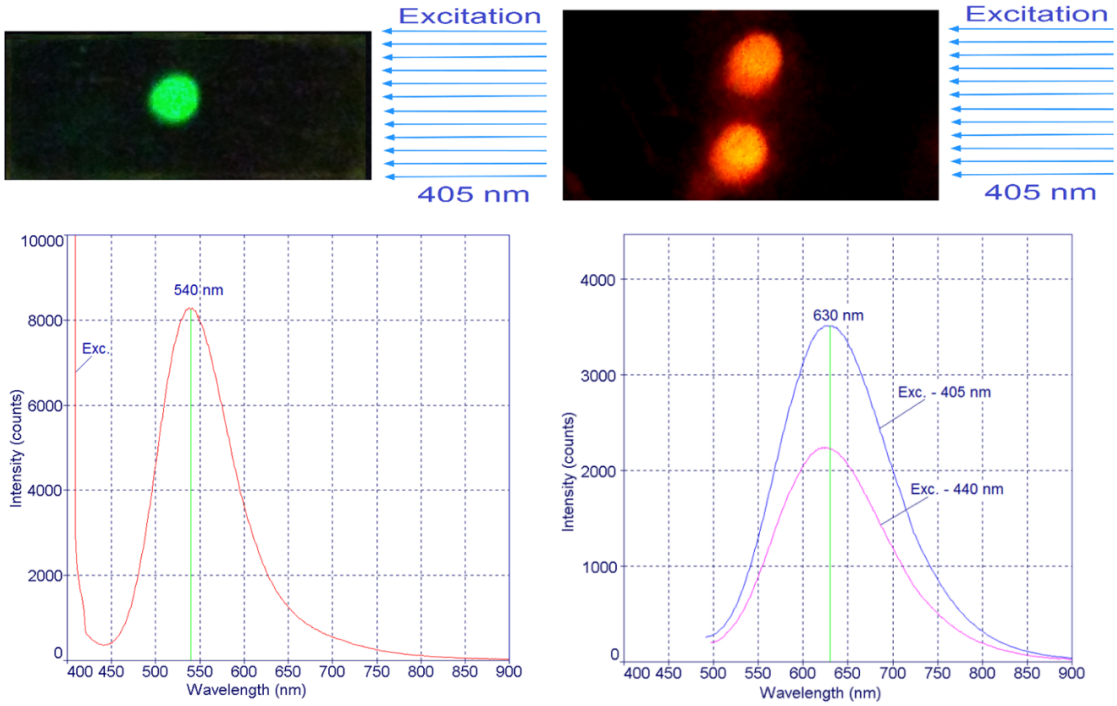

Figure 1: Photographs of exposed spots visualized with diode laser-excited fluorescent light (top). Luminescence spectra of work centers formed by femtosecond laser radiation (bottom).

The developed nonlinear photographic materials with luminescent imaging of images must meet the following requirements.

- First, these should be media in which the exciton mechanism of primary defect formation is realized. Then, electron-hole pairs and excitons generated in the course of highly nonlinear photoionization will create primary radiation defects in the medium and then their luminescent aggregates (color centers), which are formed in the processes of charge exchange, migration, and association of primary defects.

- Second, the resulting defects, primary or aggregate, must luminesce upon additional photoexcitation in order to provide luminescent visualization of the spatial distribution of the interaction of light and matter, preserved in the form of the spatial distribution of the concentrations of luminescent centers.

- Third, this luminescence should not be thermally quenched at room temperature, and the radiation defects (color centers) responsible for it should be optically and thermally stable. This makes it possible to store images formed in these environments for a long time and to visualize and examine them many times.

There are few such materials. These include crystalline compounds: LiF, NaF, MgF_2 and some others. We have developed several new crystalline media in which paint work centers are based on impurities specially introduced into these media.

Fig. 1 shows photographs of two crystal plates with different impurity ions. These plates were preliminarily exposed by a stationary femtosecond laser beam about 1 mm in diameter. As a result, color centers were formed in the irradiated regions of the plates, which were visualized under the action of radiation from a cw diode laser with a wavelength of 405 nm. The luminescence spectra of the corresponding work centers are shown below.

Volumetric photographing, storage of images, their subsequent multiple time photoluminescent visualization and detailed study by various methods of the spatial distributions of the luminescent characteristics of irradiated optical carriers provide rich information on the mechanisms of interaction between light and matter.

Such media are also used for the manufacture of optical media for direct laser recording of information in digital formats or in the form of images, including color [4].

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Laser-induced crystallization of titanium dioxide (TiO_2) nanotubular layer for photocatalytic applications

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During last two decades, TiO_2 nanotube (TNT) layers receive much attention due to their potential use in many applications, such as dye sensitized solar cells and water purification [1,2]. For such applications, the crystalline structure of material is a crucial parameter as the amorphous phase may contain various defects, which lead to recombination of photo-generated electron-hole pairs before they can be involved in a chemical reaction [3]. Among TiO_2 crystalline phases, anatase is most functional in dye-sensitized solar cells [1]. Among TiO_2 nanomaterials, anodic TNT layers are the most promising structures, which represent several µm long nanotube arrays with large surface areas.

Highly ordered anodic TNT layers can be grown in fluoride ion containing electrolytes and as-formed TNT layers are amorphous [4]. Their dimensions can be varied by adjusting the growth conditions. For their crystallization, thermal annealing is one of the widely used methods, which is energy-intensive and needs several hours of treatment at a few hundred of °C. Under thermal annealing at temperatures below 500-600°C, amorphous TNTs crystallize into the pure anatase phase [5] while at higher temperatures a mixture of anatase and rutile is formed with a higher rutile fraction at increasing the annealing temperature. Therefore, a fast energy-efficient method to crystallize TNT layers into a desired anatase phase in a non-destructive way is of high demand. In this talk, we will present the results of laser-induced crystallization of TNT layers using UV ps laser pulses. XRD and Raman spectroscopy analysis of TNT shows the anatase phase with a negligible fraction of rutile under optimal irradiation conditions. Photoelectrochemical measurement, photoelectron spectroscopy and electron microscopy results of oven and laser-annealed TNT layers will be compared.

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Formation of nanogratings on the surface of nanoporous glass under the action of femtosecond laser pulses in the visible range

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Nanosized periodic structures (nanogratings) are widely used in various micro-optical devices. The formation of nanogratings occurs under the action of ultrashort laser pulses with an intensity of ~ 10 TW/cm^2 [1] in the plane of perpendicular to polarization of laser beam [2]. The parameters of nanogratings can be controlled by varying the wavelength, duration and energy of laser pulses, as well as the repetition rate and direction of scanning. Nanoporous silicate glasses are well suited as a medium for local optical and structural modification due to the higher contrast of the refractive index and low mass density.

In this work, exposed to femtosecond (300 fs) laser pulses in the visible range (λ =515 nm) on the surface of nanoporous glass arrays of nanogratings were formed at different values of the pulse energy (E_{las} = 41–72 nJ, threshold energy E_{abl} = 65 nJ). Scanning electron microscopy shows the presence of nanogratings on the surface with a groove period of Λ = 100–150 nm. Analysis of the reflection spectra of the obtained arrays demonstrates the antireflection effect in the wavelength range of 410–440 nm. The paper also discusses the formation mechanism of nanogratings.

The study was supported by a grant from the Russian Science Foundation (project № 20-71-10103).

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Direct femtosecond laser recording in the volume of transparent dielectrics

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Femtosecond laser recording of birefringent subwavelength nanolattices in the bulk of dielectrics has been studied for almost two decades [1, 2], promising a variety of interesting applications in the field of polarizing macro-optics (see bibliography in reviews [3, 4]). The formation of subwavelength nanolattices in a plane perpendicular to the optical axis of laser radiation has long remained unexplained, and it is only in recent years that scattering processes have been considered the interaction of incident ultrashort laser pulses with cumulative multi-pulse damage to the optical uniformity of dielectrics and the interference of incident and scattered radiation with the formation of an interference pattern.

At the same time, the formation of subwavelength nanolattices as a result of nanoscale transport of chemical components of glass (mainly oxygen) and the corresponding anisotropic periodic modulation of the refractive index has so far been demonstrated only for silicate glasses [5].

In this paper, model experiments were performed to record birefringent microstructures under the action of focused ultrashort femto-picosecond laser pulses with several fixed pulse energy levels at a fixed depth in the volume of fluorite, which is a material with a Frenkel pair of point defects “interstitial-vacancy” of fluorine [6]. In particular, the experiments were performed in the volume of nanoporous glass, where the pair is oxygen and silicon. The phase shift is measured for various conditions of recording microstructures, and as a result of this analysis, a mechanism is proposed that explains the formation of nanolattices responsible for birefringence in the fluorite volume, their location and orientation with respect to the optical axis of the recording laser radiation, as well as the nature of the dependence of the phase shift on the energy/intensity of laser pulses.

The study is funded by the grant of Russian Science Foundation (project № 20-71-10103).

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Processing of arrays of microchannels in fused silica by picosecond laser ablation

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Ultrashort laser pulses have demonstrated their interests for ablation and structuration of transparent dielectric materials at the micrometric scale. Spatially shaping the intensity distribution of the laser irradiation therefore enables to broaden the range of patterns accessible with single-shot laser ablation.

Using a micro-Bessel beam generated with an axicon, we first demonstrate the realization of single microchannels at the front surface of fused silica. Blind channels of few tens of micrometres long and $\sim$ 1 µm diameter are obtained upon single-shot, 1-ps, 800-nm laser irradiation. These channels exhibit a good quality of their entrance, without rims or recast material [1]. We further implement additional amplitude filtering with an annular aperture, that provides a straightforward technique to offer length-tunability to the process. This way, we obtain few-micrometres-long microchannels.

Then, in view of realization of micro-components based on arrays of microchannels, we investigate the possibility to process patterns composed of dense meshgrid of short-length channels [2]. We address experimentally the crucial problem of crosstalk, i.e. the influence of the preexistence of the previous channel on the propagation of the laser beam and, in turn, on the processing of the next one. We investigate how this can yield severe limitation to the process when the spatial pitch between adjacent channels approaches the micrometre range, despite the robustness of Bessel beam to reform after obstacles. Finally, we identify a regime within which arrays of submicron-diameter channels with variable lengths can be processed one-by-one with a spatial periodicity down to 1.5 µm.

This is a first step towards the realization of microscale integrated optical components by direct laser ablation, as an alternative to multistep technologies, offering a vacuum-free, nonchemical, flexible, programmable, fast and affordable technique.

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Femtosecond laser writing of hollow channels in nanoporous silicate matrix

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Microfluidics is a rapidly expanding field as it provides the ability to miniaturize and integrate systems for chemical and biological analysis [1]. It brings compact devices that handle small amounts of liquid (pico/nanoliter volumes) [2]. A great inconvenience for the development of microfluidics is now the availability of technologies for recording channels and systems with the desired parameters. Direct laser writing can compete with most other technologies (printing, photolithography in polymers, etc. [3]) if it demonstrates the possibility of creating channels with adjustable parameters without toxic etchants. In this work, we demonstrate the results of the fabrication of buried channels in a nanoporous silicate matrix. Subsequent ultrasonic cleaning of the sample in water results in hollow channel formation.

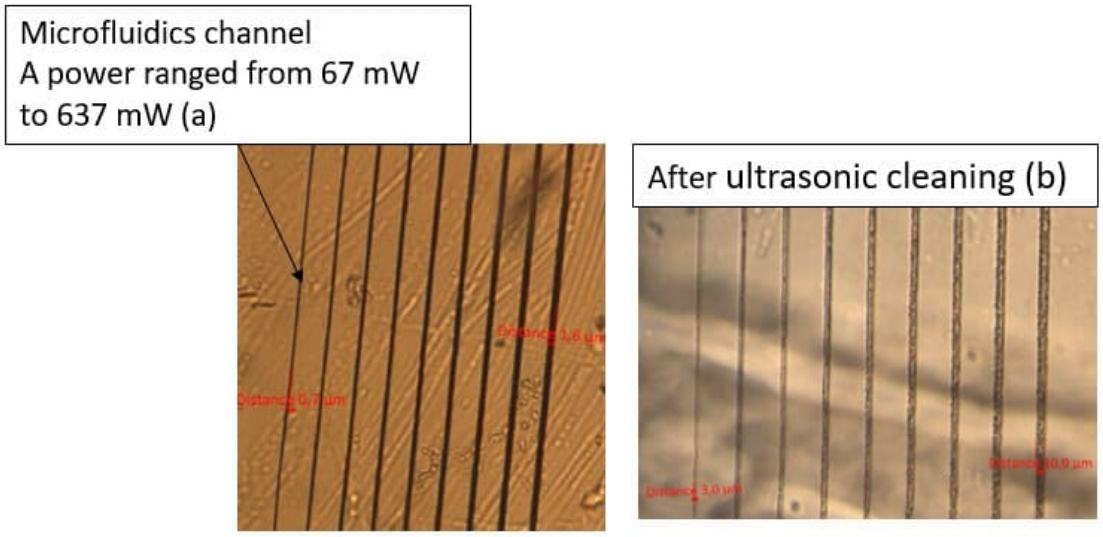

Figure 1: Microphoto of channels after writing (a), ultrasonic cleaning (b), and schematic view of the system

We propose laser writing inside of nanoporous silicates matrices (NPSM-17) in the form of the plane-parallel plate with the free volume of pores 25%. For this, a femtosecond laser (Avesta, Antaus) operating at 1035 nm wavelength (220 fs, up to 1 MHz) was utilized. As a result, a set of channels with minimal cross-sections $0.7-2.0$ µm were fabricated (Figure 1, a). After the cleaning steps, the recorded structures were freed of degraded material, providing empty space in the channel section (Figure 1, b). Additional heat treatment made it possible to significantly change the roughness of the channels.

The study is funded by the grant of Russian Science Foundation (project № 20-71-10103).

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Microstructure and droplet formation via extreme energy deposition of 3 µm laser radiation in water

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One of the unique properties of laser radiation at a wavelength of about 3 µm is a strong fundamental absorption line ($1.3 \cdot 10^4$ cm^{-1}) in water. It allows laser sources operating at this wavelength to be used for transparent materials microstructuring including semiconductors via laser-induced backside wet etching (LIBWE) and 3D bioprinting with laser-induced forward transfer (LIFT) method.

In recent years the microstructuring techniques involving the use of a buffer liquid proved to be very effective, as such liquid can increase the accuracy and controllability of the processing method. One of these methods is LIBWE, which is used for transparent materials processing. Compared to other conventional methods, it is characterized by a low threshold, low etching rate, and a high-quality surface with very low surface roughness. Using this method submicron and nanoscale structures can be formed on the processed sample surface. Usually, in this technique nanosecond UV-NIR lasers are used in combination with strongly absorbing media, for example, organic dyes or liquid metal. Extreme absorption of 3-µm radiation in water makes laser sources at this wavelength attractive for many applications where a high energy input into the liquid is required [1]. Up to now, the etching process at high energy deposition is not understood. It includes a combination of complex physicochemical processes in water, including nonlinear bleaching, sharp temperature and pressure increase, as well as strong mechanical effects, such as cavitation and shock wave generation.

Another interesting application of nanosecond 3-µm laser radiation involving buffer fluid is the 3D bioprinting with laser-induced forward transfer (LIFT) technique. In this method, bioink microdroplets are formed due to the interaction of laser radiation with hydrogel. LIFT bioprinting is characterized by acceptable resolution and print speed for most applications and also high cell survivability [2]. However, droplet formation is a complicated hydrodynamical process and by studying it, precise and controllable biological microstructures can be formed.

We report the study of the surface structuring of crystalline materials during LIBWE in water under the excitation of powerful 3-µm laser pulses. The dependence of microstructure morphology on the irradiation regime is discussed. Depending on the laser radiation influence mode in the etching process, both acoustic effects (cavitation and shock waves generation) and active chemical reactions, accompanied by the formation of precipitate, can play a decisive role. A method for increasing the etching rate by using an alkali solution is considered. Also, different jetting regimes during droplet formation in LIFT with different laser fluences are presented and the properties of the obtained biostructures are discussed.

The part of the work dedicated to laser radiation influence on water was supported by Russian Scientific Foundation (RSF) (17-72-20130). The development of the LIFT method was supported by RSF grant 20-14-00286.

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Nanostructuring of materials for photonics with designer ultrafast laser pulses

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The capability to restrict energy deposition is key in designing accurate laser processing technologies and in fostering progress in laser-based micro/nano-technologies. Optical resolution being limited by diffraction, new methods are required to harvest either near-field effects or to build up on material reactions, restricting the processing scale well below the diffraction limit. This excitation confinement, along with the nonlinearity of the interaction between ultrashort laser pulses and dielectric materials, is at the base of refractive index engineering and forms the building block for developing volume embedded optical functions. We will explore the potential of designing the beams in space and time to achieve interaction control and high resolution in three dimensions and to fabricate complex hybrid micro-nano optical systems, capable of transporting, manipulating and accessing optical signals. We indicate a range of applications, from telecom to astrophotonics, including applications designed for the mid-IR spectral range for sensing and imaging.

3D microfluidic SERS chips fabricated by hybrid femtosecond laser processing for attomolar sensing

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We have developed hybrid femtosecond laser processing for fabrication of 3D microfluidic SERS chips enabling real-time sensing with ultrahigh sensitivity [1]. In this process (Fig. 1), 3D glass microfluidic channels are first fabricated by femtosecond-laser-assisted wet etching. This is followed by the space-selective formation of Ag thin films inside the microfluidic channels via femtosecond laser direct writing ablation and electroless metal plating. The Ag films are subsequently nanostructured by irradiation with linearly polarized femtosecond laser to form periodic surface nanostructures. The resulting microfluidic SERS chip exhibited an enhancement factor of 7.3×10^8, and then capability of the real-time detection of Cd^{2+} ions with different concentrations from 10 ppb to 10 ppm. We have further proposed a novel strategy by taking advantage of the microfluidic configuration to achieve the detection limit down to aM [2]. The microfluidic chip can produce an interface of analyte solution and air on the SERS substrate in the microfluidic channel, which locally aggregates the analyte molecules at the interface during the measurements (liquid-interface assisted SERS, LI-SERS). The LI-SERS technique achieves the enhancement factor reaching 1.5×10^{14} with a detection limit below 10 aM. The mechanism of extraordinary enhancement by LI-SERS might be attributed to the Marangoni convection induced by photothermal effect. We believe that the LI-SERS technique is versatile and can be used for trace detection even for substances with extremely low concentration including the screening of viruses such as COVID-19 without DNA amplification.

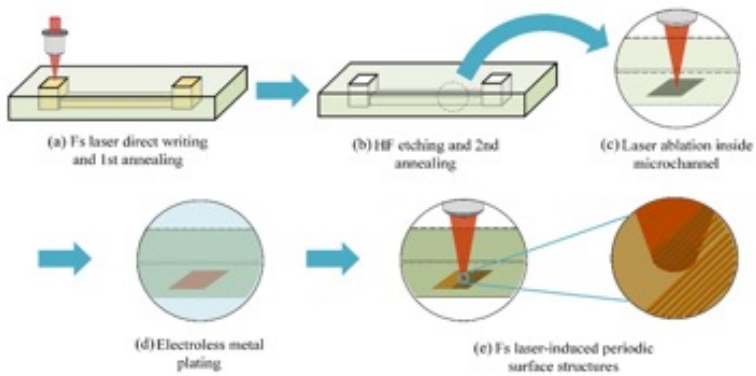

Figure 1: Hybrid femtosecond laser processing for fabrication of 3D microfluidic SERS chips.

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Micro-modification of optical properties in cadmium sulfide-doped silicate glass by a femtosecond laser beam

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Modification of the glass structure with a femtosecond laser beam makes it possible to initiate the processes of structural rearrangements and local changes in properties of the material at the nano- and microscale [1]. In this regard, oxide glasses doped with semiconductors are promising objects for femtosecond laser processing. The great interest to these glasses is caused by their unique optical properties determined by the size of semiconductor quantum dots. Their growth inside glass can be induced by additional heat treatment at temperatures near the glass transition. On the other hand, direct laser writing used for space-selective precipitation of quantum dots in oxide glasses is a promising way to the development of integrated waveguides, photonic integrated circuits and optical data storage based on luminescence of the laser-written microdomains [2,3].

Here we report results on the direct laser writing of the domains in Na_2O-ZnO-B_2O_3-SiO_2 glass doped with 4 wt.% CdS. Femtosecond Yb:KGW laser Pharos SP (Light Conversion Ltd.) emitting ultrashort pulses at 1030 ± 2 nm central wavelength was used for direct laser writing. The laser was tuned at 100 kHz pulse repetition rate and $\sim$ 180 fs pulse duration. The number of deposited laser pulses was varied from 10^1 to 10^6. Laser pulses were focused at a depth of $\sim$ 150 µm under the glass sample surface with Olympus microscope objective (20X, N.A.=0.45). A half-wave plate was used for the rotation of laser beam polarization at 0, 45 and 90°.

Laser processing of $10 \times 10 \times 3$ mm polished plane-parallel glass sample by the stationary beam induced pipe-shaped domains from 1.5 to 16 µm in diameter. Irradiation by 10^3–10^6 laser pulses produces the yellow coloration of domains which can be attributed to CdS quantum dots precipitation. Domains written by fewer pulses were colorless. Under 400–410 nm excitation selected with an interference filter from the mercury lamp light, domains manifested luminescence in the visible spectral range. Polarized light microscopy revealed optical retardance for the domains written at 10^4–10^6 laser pulses. Presumably, the nature of this phenomenon may be associated with linear dichroism or intrinsic birefringence of oriented CdS nanocrystals. For each modification area, maximum luminescence intensity and retardance were confined to the ring-shaped periphery area of the microdomains. This can be directly related to the Gaussian profile of the laser beam, which is decisive for laser-induced temperature field distribution and CdS quantum dots precipitation. At the same time, the variation in the number and energy of laser pulses was found to contribute to the changes in the coloration and luminescence intensity as well as to the retardance value. This is most likely due to the variation of the thermal conditions in the laser modification area and, consequently, of the size of the precipitated CdS quantum dots.

This study was financially supported by the Council for Grants of the President of Russian Federation (grant MK-1497.2020.3) and the Russian Foundation for Basic Research (grant 21-53-12026).

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Femtosecond laser writing of waveguides inside nanoporous silicate matrix

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To support the growing field in waveguide sensoric [1] we demonstrate buried waveguides fabrication by femtosecond laser pulses, that have achieved high sensitivity to external molecules. Recently, portable devices with such a nanoporous glass loaded with organic indicators were demonstrated for monitoring the level of formaldehyde, nitrogen dioxide, and ozone [2]. The interaction of the indicator and the harmful gases changes the color of the entire glass plate. However, information processing was performed by the spectral analysis of the glass plate. For a more accurate analysis of chemical reactions in such a nanoporous medium, we suggest inscribing chip-scale optical waveguides.

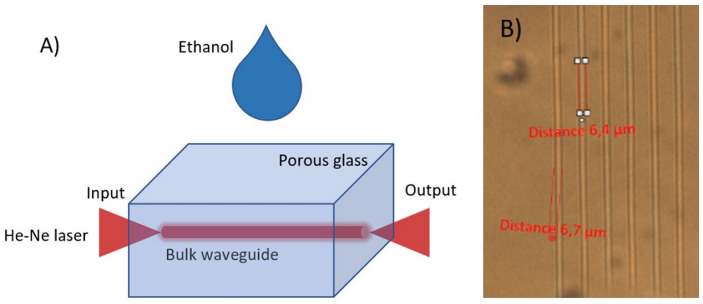

Figure 1: Schematical view of the barrier and microphoto of the build barrier

We propose laser writing inside of nanoporous silicates matrices (NPSM-17) in the form of the plane-parallel plate ($20 \times 20 \times 1$ mm) with the free volume of pores 25%. For this, a femtosecond laser (Avesta, Antaus) operating at 1035 nm wavelength with a pulse duration of 220 fs and repetition rate up to 1 MHz was utilized. As a result, we obtained the processing parameters to achieve waveguides with aspect ratio 1:1 operating in a single-mode regime (Figure, a). The sensor operation is based on the influence of target molecules on the propagated optical signal. Thus, the presence of ethanol molecules changes the refractive index contrast for $2 \cdot 10^{-4}$. The results show the optical sensitivity of waveguides inscribed in PG for the detection of small molecules such as ethanol.

The study is funded by the grant of Russian Science Foundation (project № 20-71-10103)

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Barriers building inside nanoporous silicate matrix by femtosecond laser pulses

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Nanoporous silicate optical media is a promising platform for molecular and biological research [1]. It has already been repeatedly stated about the integration of photonic and fluid [2] micro-sized components into similar matrices, which aim to transform the site into an all-optics and chips-scale device. Another important component is physico-chemical separation, which allows the organization of many closed sectors in the glass plate. In another word, it's a "barrier", which is formed in the area of action of sharply focused femtosecond laser pulses as a result of a change in the material's density [3]. In this work, we carry out a study to optimize the formation of barriers and have determined the threshold recording values and their geometric dimensions.

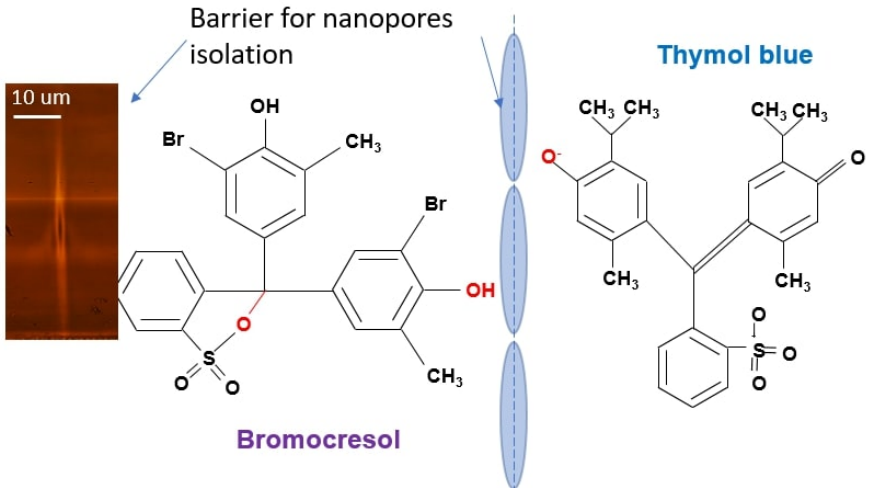

Figure 1: Schematical view of the barrier and microphoto of the build barrier

We propose LDW inside of nanoporous silicates matrices (NPSM-17) in the form of the plane-parallel plate (20×20×1 mm) with the free volume of pores 25%. For this, a femtosecond laser (Avesta, Antaus) operating at 1035 nm wavelength with a pulse duration of 220 fs and repetition rate up to 1 MHz was utilized. As a result, we obtained the processing parameters to build a set of barriers, investigated its structural properties (Figure), and formed a few isolated sectors inside the nanoporous matrix. The created cells were also filled with various fluorescent dyes.

The study is funded by the grant of Russian Science Foundation (project № 20-71-10103).

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Femtosecond structuring of nanoporous silicate matrices

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Femtosecond laser writing of three-dimensional structures in glass is a key technology for fabrication of compact (chip scale) devices - sensors, photonic chips, biomedical platforms. Various structural modification depending on the intensity of laser radiation, can be formed [1] - an increase in material density, generation of point defects, nanogratings, and material decompaction. The selection of nanoporous silicate matrices [2] expands the thresholds for laser processing, the functionality of structures and thus suggests the novel structures fabrication.

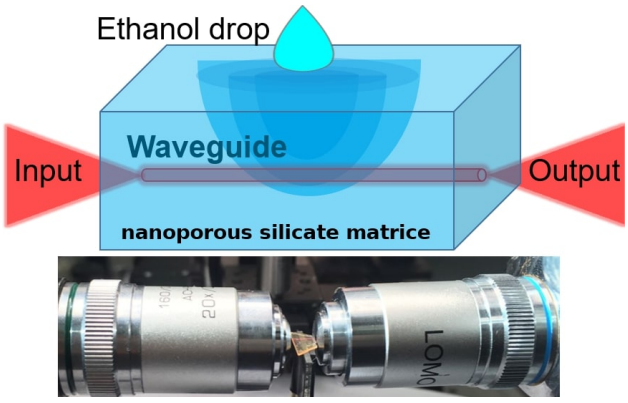

Figure 1: Illustration of waveguide sensor element in porous glass operation (on top). Experimental investigation of the sensor (below)

We propose LDW inside of a nanoporous silicates matrices (NPSM-17) in the form of plane-parallel plate ($20 \times 20 \times 1$ mm) with free volume of pores 25%. For this a femtosecond laser (Avesta, Antaus) operating at 1035 nm wavelength with a pulse duration of 220 fs and repetition rate up to 1 MHz was utilized. As a results, a set of structures were fabricated - waveguides of several types, microfluidic channels and barriers. We considered the application of some structures in the manufacture of waveguide sensors for liquids and gas components (Figure).

The study is funded by the grant of Russian Science Foundation (project № 20-71-10103)

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Mini-simposium “Diamond Photonics”

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Scope

Formation of NV centers in diamond

Microctructuring of bulk diamond

Application of color centers in diamond

Excitation of luminescence of color centers in diamonds

Femtosecond optical breakdown of diamond

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Theoretical calculation on transient electronic and optical properties of the photo excited diamond, irradiated by intense femtosecond laser pulses with visible wavelengths is reported. For low intensities, photoinoziation of diamond follows the predictions of the perturbative non-linear optics, a qualitative change of the electron dynamics occurs when the laser intensity increases above $5 \cdot 10^{14}$ W/cm^2. We find that in this high intensity regime, laser energy absorption is enhanced due to excitation of bulk plasmon resonance. The electron density difference distribution shows the effect of softening of C–C bonds resulting in instability of the diamond lattice.

Ultrafast laser writing for integrated quantum diamond technology

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Diamond has created an interest in the quantum technologies community due to a naturally occurring imperfection, the Nitrogen Vacancy (NV) color center. The NV center has emerged as a candidate for quantum computing and field sensing due to its long electron spin coherence times and its ability to be found, manipulated and read out optically [1]. Additionally, the spin states are magnetic and electric field sensitive due to Zeeman and Stark effects, respectively and hence can be utilized as atom sized sensors for field sensing applications.

In this work, we will demonstrate ultrafast laser writing as a powerful platform for integrated quantum photonics in diamond. In the recent years, laser writing has allowed 3-dimensional formation of photonic and microfluidic devices in diamond [2]. It has shown tremendous capabilities, especially in the ability to integrate laser written waveguide with a deterministically positioned NV center, utilizing the same laser writing setup, as shown in Fig. 1(a). Additionally, laser writing in CVD diamond with dense Nitrogen concentrations has enabled the formation of high-density NV ensembles within the waveguides, as shown in Fig 1(b). For field sensing applications, ensemble NVs will be beneficial as they allow high sensitivities of detection. We will discuss ways to further improve the sensitivities of photonic networks and integrate with other laser written elements for fully integrated lab-on-chip devices.

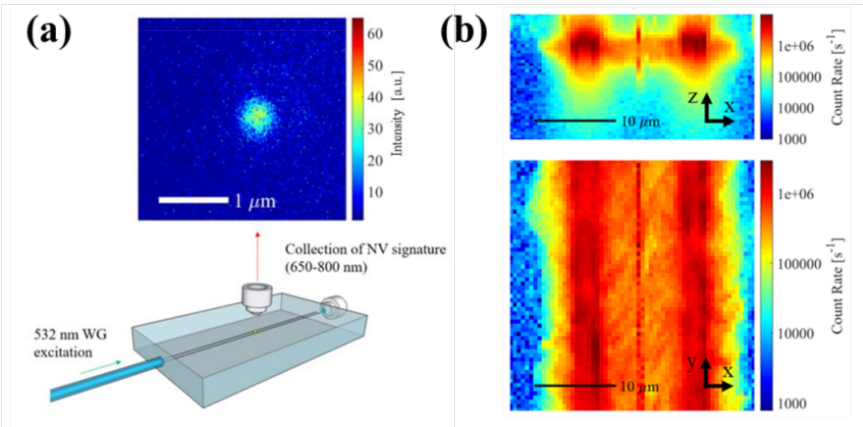

Figure 1: (a) Schematic showing 532 nm excitation of a laser inscribed single NV coupled to a laser written waveguide. EMCCD image of the NV emission is shown above the schematic. (b) Overhead (below) and cross-sectional (above) Photoluminescence maps from the laser inscribed NVs within waveguides in high Nitrogen concentration HPHT diamond.

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Laser-induced luminescent centers in diamond: influence of exposure and duration of short laser pulses

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Ultrashort-laser modification of atomistic optical centers in diamonds is one of the most interesting research areas with many real-life applications includes sensing, quantum communication and "stealth" luminescent micro-marking of natural and synthetic diamonds for their legal tracing [1-3].

In this work, we report on pulsed laser induced generation of luminescent centers in diamond without surface and volume graphitization. Laser recording was carried out in the volume of natural and synthetic diamonds at a wavelength of 515 nm with different intensities and exposure times. Each series (matrix) was recorded at pulse durations of 300 fs, 1 ps, and 3 ps, respectively.

Visualization and analysis of the luminescence of the modified regions was carried out using a scanning laser Raman microscope (Confotec® MR350) with a pumping wavelength of 532 nm. An example of a matrix is shown in Fig. 1.

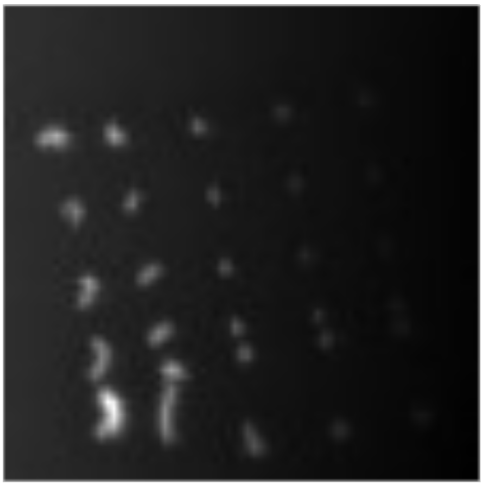

Figure 1: The matrix of laser-induced luminescent centers in diamond. Image obtained on a scanning laser Raman microscope (pump 532 nm).

This work was funded by the grant of Russian Science Foundation (project № 21-79-30063).

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Effect of convergent beam aberrations on laser focusing in dielectrics

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Focusing of ultrashort laser pulses is widely used for laser processing and creation of microstructures inside dielectric materials. Laser energy is mainly absorbed in focal spot due to nonlinear features of laser-dielectric interaction [1]. The size of focused laser spot depends on pulse duration, pulse energy, and focusing lens numerical aperture [2].

In this paper, the influence of numerical aperture on focal spot size is investigated. During the experimental studies radiation of femtosecond laser Satsuma (1030 nm, 0.3-10 ps) was focused by microscope objectives with different numerical apertures on front and back surfaces of samples made of ZnSe and diamond. The conducted studies have shown that with an increase in numerical aperture when convergent laser beams pass the air-dielectric boundary aberrations leading to an increase in focal spot size occur. Therefore, when calculating focal spot size both laser-dielectric interaction effects and aberrations should be taken into account.

This study is funded by the grant of Russian Science Foundation (project no. 21-79-30063).

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Ultrafast nonthermal formation of NV centers in diamond

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Recent studies have shown that color centers like NV centers in diamond are promising candidates for technological applications, e.g. quantum sensing, secure message encryption and biological imaging. They can also be used as Qubits. In order to construct arrays of NV centers for quantum computing, it is necessary to maximize interqubit interactions and, at the same time, minimize all sources of decoherence. Unfortunately, the fabrication process of NV centers by ion bombardement or artificial growth is not sufficiently controllable and comes with a high increase of the crystal temperature. Due to the heating process, other defects could appear within the lattice surrounding that could decrease the efficiency of the NV center. Therefore, we simulated the formation of a NV center driven by femtosecond-laser excitation by ab-initio methods. For this, we first performed ab initio molecular dynamics simulations of diamond with a defect density of 3.1% and analyzed the impact on ultrafast phenomena like thermal phonon squeezing. We used this knowledge to perform simulations of a supercell containing a nitrogen atom and a vacancy as not nearest neighbors initially. Our results indicate, that femtosecond-laser pulses could be used to controllably produce NV centers nonthermally within 250 fs. Instead of remaining near a grid point, like in the thermal ground state, the nitrogen atom starts to oscillate between two grid points after the ultrashort-laser excitation. For comparison to the thermal case we performed a single run in which a NV center is thermally formed at T = 1500 K in one of our supercells [1].

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Photolytic formation of NV complexes in diamond exposed to UV femtosecond pulses

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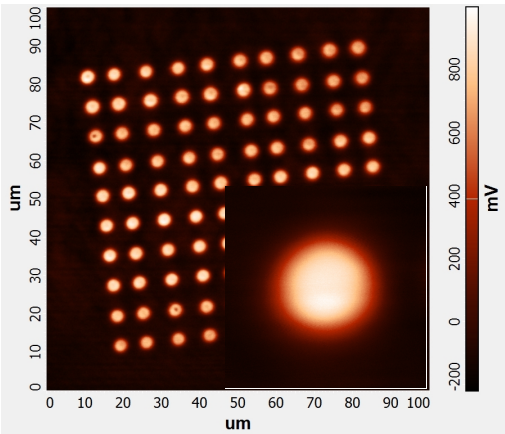
Laser-induced formation of nitrogen-vacancy defects in a chemical vapor deposition (CVD)-grown diamond monocrystal is reported in the preablation regime, when slow surface etching ($< 10^{-4}$ nm/pulse), typically called nanoablation [1], occurs. The experiments prove that besides the well-known process of graphitization [2], which leads to a complete collapse of diamond lattice, laser-induced electronic excitation enables the photolytic formation of defects - a bond rearrangement of the point type.

Luminescence measurements confirm that during irradiation by 266-nm femtosecond pulses, the nitrogen-vacancy concentration grows logarithmically and can increase tenfold until it reaches saturation. This process is shown to be accompanied by a progressive decrease in surface reflectance attributed to Frenkel pair generation. Neither surface ablation nor diamond graphitization in the irradiated zone are found. The relationship between the coloration of the diamond and the nanoablation of its surface as well as the possible mechanisms of the photolytic reconfiguration of bonds in the diamond are discussed.

The results indicate femtosecond laser nanoablation to be a promising tool for a precise control of the number of produced vacancies in the lattice and, hence, for managing the probability of formation of an individual NV center in the desired point of a crystal. The possible routine of laser processing providing controlled deterministic creation of a single NV center is proposed. Besides practical significance, this study gives new insight into laser-induced processes in diamond, considering the point defect creation as a preliminary stage of accumulative graphitization and nanoablation.



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Ultrafast polarization resolved surface and bulk ionization effects in natural diamond

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The market of the synthetic diamonds is expanding every year with unimaginable speed. That is why the mapping and tracing of each diamond is relevant nowadays to showcase its origin. Over the years, many methods have been developed for mapping both natural and synthetic diamonds. However, the optical methods, such as IR spectroscopy, Raman spectroscopy, luminescence and others, remain as the most informative methods among them. Highly sensitive photoluminescence method [1, 2] is used to for study the fine spectrum of diamond optical centers, and one of the advantages of such methods is a fairly wide spectrum of excited emission.

In our studies, we demonstrated the dependence of the surface (surface ablation process) and bulk ionization (photoluminescence excitation) effects on the state of polarization of the exciting radiation. Ultrashort pulses from the ytterbium laser Satsuma with wavelengths 515 nm and 1030 nm were used to excite different effects in the diamond slabs of different crystallographic planes, namely (100), (110) and (111). The duration of ultrashort pulses used in studies varied from 300 fs to 3 ps with repetition rate the dependences were obtained. For surface effects polarization resolved studies showed the change in the threshold fluence of the ablation as well as the minimum diameter of the craters. Additionally, such studies can allow one to determine the crystallographic planes of the diamond without the need of the sophisticated devices. For photoluminescence studies the dependence of the photoluminescence yield on the state of the polarization was obtained. The obtained results will allow to expand the already existing techniques of natural diamond tracing. Additionally, the polarization photoluminescence studies can complement already existing diamond mapping techniques and allow more precise determination of the defects and optical centers.

This work was supported by the Russian Science Foundation (Grant No. 21-79-30063).

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Overview of basic processes underlying ultrafast intra-center and interband photoexcitations in bulk diamond for micromarking and tracing applications

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This work presents an overview of basic fundamental processes, underlying ultrafast optical and electronic dynamics during femtosecond-laser photoexcitations in natural diamonds (fig. 1a) for micromarking and tracing applications. A number of consequent process — non-linear laser propagation of femtosecond-laser pulses [1], spectrally- and intensity-dependent non-linear photoionization mechanisms [2], Auger recombination and ambipolar diffusion of dense electron-hole plasma, generation of Frenkel "interstitial-vacancy" pair, atomistic processes of laser-induced modification of the present optical centers in diamond are overviewed, using our recent original results.

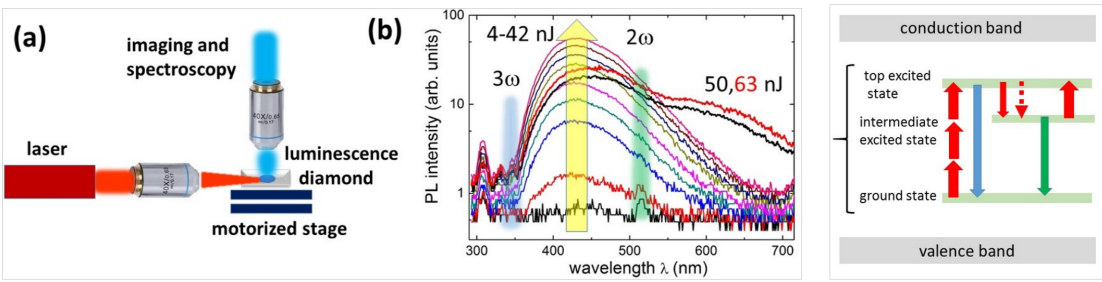

Figure 1: (a) Schematic of 1030-nm high-NA femtosecond-laser photoexcitation in diamond. (b) 3-photon blue-range A-band intra-center photoluminescence dynamically switching to 2-photon green-range photoluminescence from the intermediate process due to upper-level saturation, and the related three-level energy diagram.

This study is funded by the grant of Russian Science Foundation (project no. 21-79-30063).

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Linear and nonlinear excitation of the A-band of luminescence in diamond

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In the luminescence spectra of natural and synthetic diamonds, a broad luminescence band is often observed, occupying the region from ∼350–700 nm, the maximum of which is near ∼480 nm and shifts within certain limits from sample to sample [1-4]. This luminescence is excited by X-rays, accelerated electrons, and optical radiation. This band is dominant in the X-ray luminescence of most natural diamonds; therefore, it plays an important role in the technologies of luminescent separation of diamond-bearing ores. In some types of separators, diamonds are separated from other luminescent minerals taking into account the value of the decay time of the A-luminescence band [5-7]. It is known that the value of the decay time of the main time component of the luminescence also varies from sample to sample within ∼(3-9) ms [2-4]. The scatter in the spectral and temporal characteristics of the A luminescence band is due to several reasons. First, the spectra are distorted due to the reabsorption of luminescence by other centers contained in the crystals. Second, the difference in spectra is due to the presence of additional luminescence bands of centers overlapping with the A band, for example, such as H3 centers. The variability of the decay time for different samples is associated with the quenching of the A-band by extraneous centers. Considering the great importance of this luminescence for practical applications, it is necessary to know well its characteristics and be able to confidently identify it. In addition, new opportunities for the practical use of diamonds are emerging. Therefore, studies of the A-luminescence band continue [7-9].

Obviously, in crystals with the maximum decay time of the long-term luminescence component (∼9 ms), quenching by impurities is minimal. Therefore, in such crystals, the A-band has the least distorted characteristics, and its intensity is maximal. Consequently, the A-band of luminescence is the purest in such a crystals.

For such samples, the spectral, kinetic, and temperature characteristics of luminescence are studied under X-ray excitation, under linear optical interband and intracenter excitation, and also under femtosecond laser nonlinear anti-Stokes excitation. The characteristic features of the A-luminescence band have been determined, which make it possible to identify this band among the numerous other luminescence bands of diamond. The kinetics of luminescence in this band, which is identical under X-ray and optical excitation, is especially characteristic. This kinetics has reference features characteristic of the A-luminescence band. These include sharp thermal quenching in the region above ∼410 K, temperature independence of the long-term component decay time of luminescence in the region of ∼(350–170) K, and its transformation into two other components at temperatures below ∼150 K with their own characteristic temperature dependences.

According to our data, the A-band of luminescence under anti-Stokes excitation by femtosecond laser emission arises by the mechanism of nonlinear interband photoionization with subsequent recombination excitation of centers emitting the A luminescence band. This is confirmed by the coincidence of the luminescence characteristics under femtosecond excitation, under X-ray excitation, and under optical interband excitation.

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Liquid immersion for imaging defects in diamonds

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The investigation of minerals with high refractive index, such as diamond is of special interest. But scanning refractive objects of arbitrary shape is difficult due to their refractive index. The technology of immersion diamond research is innovative and effective for industrial implementation. Due to matters with refractive index similar with crystal index identification of microscopic defects not noticeable at an optical research of a crystal in the air environment is possible [1].

In this work, we consider a producing of liquid immersion based on nanoparticles with a refractive index close to that of diamond for the visible and IR ranges for broadband spectral identification, optical microscopic inspection, and laser modification of complexes of optically active point defects in the bulk of diamonds.

This work was supported by Russian Science Foundation (grant # 21-79-30063).

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Laser-induced luminescence of boron-doped synthetic diamond at various laser pulse durations

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Photoluminescence of a optically-active centers in crystalline diamond emerges as a key enabling process to realize on this platform new functional modalities for quantum technologies, sensing and "stealth" luminescent micro-marking of natural and synthetic diamonds for their legal tracing [1-3].

Photoluminescence spectra were obtained upon excitation by ultrashort laser pulses in the volume of boron doped synthetic diamond at wavelengths of 1030 nm and 515 nm with different intensities at pulse durations of 300 fs, 1,2 ps, and 6.2 ps. As a result, the dependence of photoluminescence was established for various pulse durations and power.

This work was funded by the grant of Russian Science Foundation (project № 21-79-30063).

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Chalcogenide immersion media for diamond research

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Recently, there has been an increasing interest in chalcogenide glasses [1-3], since they are promising optical materials due to their unique nonlinear optical properties. The existence of a wide range of possible glass-forming systems with a large spatial composition and good resistance to crystallization makes it possible to obtain glasses with such optical properties as nonlinearity, light sensitivity, high optical transparency in the IR range, and good photosensitivity, which can be optimized for photonic applications.

Chalcogenide glasses are one of the most widespread families of amorphous materials and are widely used in holography [4]. Chalcogenide materials must contain one or more chalcogenide elements. Pure S, Se and Te are not used due to their short service life and low sensitivity. By varying the stoichiometric ratio of chalcogenide compositions, the refractive index can be varied as shown in [5]. As is known from [6, 7], the optical properties of chalcogenide solid solutions are influenced not only by the composition, but also by the preparation conditions, thickness, and heat treatment processes.

In this work, the interaction of femtosecond laser radiation with all the most typical representatives of amorphous chalcogenides, including various stoichiometric ratios, is considered. The dependence of the transmission of glasses on the pulse energy has been established. The thresholds of damage to glasses under the action of femtosecond laser pulses of variable frequency in the near infrared range were determined. The main goal of our work is to search for optimal compositions with a high damage threshold when interacting with femtosecond laser pulses.

The study is funded by the grant of Russian Science Foundation (project № 21-79-30063).

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Craters morphology on a diamond surface after 0.3 ps and 10 ps laser ablation

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Laser ablation is used for the treatment of different materials. The material ablation, which depends on the duration of the pulse of the incident radiation, is one of the key parameters of laser processing [1]. In the course of the experiment, laser ablation of the diamond surface in air with a variable pulse duration was carried out. The morphology of the diamond surface after laser ablation of 0.3 ps and 10 ps pulse duration were experimentally obtained.

The source of laser radiation was a Satsuma fiber laser (Amplitude Systemes) with an active medium on Yb^{+3} ions (wavelength of main harmonic: 1030 nm, frequency doubling: 515 nm, spectral full width at half maximum (FWHM): 9 nm , repetition rate 0–2 MHz). The pulse duration at half maximum τ_p was smoothly varied for infra red fundamental pulses using an output grating compressor in the range of 0.3—10 ps. The radiation was focused by a lens with a numerical apertures 0.65. The crater profile was obtained using a scanning probe microscope. Then, the dependencies of the crater depth on the energy density for different pulse durations were plotted.

This work was supported by Russian Science Foundation (grant # 21-79-30063).

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Spectral transformation of visible and IR ultrashort laser pulses in polycrystalline ZnSe

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Advances in the production of synthetic diamonds, leading to a decrease in their cost and an increase in production volumes, as well as the problem of counterfeit diamonds, require the development of more rigorous methods for marking and identifying natural diamonds. One of the methods for solving this problem is the use of immersion media, which will make it possible to identify uncut diamonds, study its point defects and carry out modification in its volume with laser radiation. The immersion medium should have a refractive index close to the refractive index of diamond (2.40–2.46 in the visible range of the spectrum) for the visible and near-IR ranges, in which all absorption and luminescence bands of point defects in diamond are located. High index chalcogenide glasses meet this requirement. Also, chalcogenide glasses soften at temperatures from 70 to 500 C, which will allow diamond to be fused into them without strong overheating. [1].

In this work, a series of experiments was carried out to measure the spectral transformation of ultrashort pulses in the IR (1030 nm) and visible (515 nm) ranges at different pulse energies and pulse durations: 300 fs, 1 ps, and 10 ps when focusing into the volume with a lens with a numerical aperture NA = 0.25 in polycrystalline ZnSe. Different values of broadening for the red and blue shoulders of the pulses are found, which are related to the self-phase modulation of the leading and trailing edges of the pulse, respectively. The results are discussed [2].

Studies were funded by the grant of Russian Science Foundation (project № 21-79-30063).

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Efficient NV center formation inside diamond by femtosecond laser pulses

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Nitrogen-vacancy (NV) center in diamond has attractive attention as a platform of quantum applications ranging from computing [1] to sensing [2]. The formation of the NV center by the femtosecond laser irradiation followed by thermal treatment has been recently reported [3]. Based on the photoluminescence observation, the formation mechanism of NV center by femtosecond-laser irradiation has been suggested that a substitutional nitrogen in the vicinity of focus was converted into a single NV- center due to recombination with a photo-generated neutral vacancy (GR1 centers). The remarkable benefit of such femtosecond laser direct-writing method is to realize flexible spatial arrangement of NV center in three-dimension. This is unable to be achieved by other methods using electron or ion beams. Previously we have observed the efficient graphitization in diamond by the femtosecond double-pulses irradiation compared to the conventional single beam irradiation [4]. Here we report the efficient NV center formation by the femtosecond laser pulses.

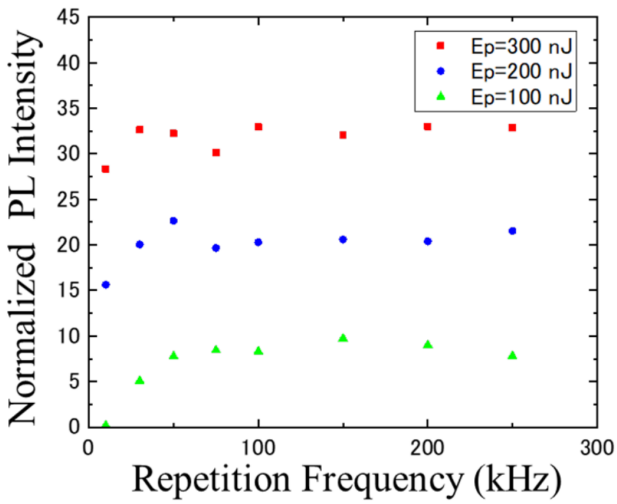

Figure 1: PL intensity from NV centers in diamond as a function of pulse repetition rate.

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Formation of luminescent defects in artificial diamonds by femtosecond laser pulses

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Today, diamonds are the most expensive material for jewelry decoration. At the same time, the share of artificial diamonds in the jewelry market is growing and the number of companies engaged in the synthesis of diamonds for commercial purposes is also increasing. In this regard, the problem of identifying diamonds arises not only according to the natural – artificial principle, but also in which company this or that diamond was synthesized. Competition appears within the artificial diamond market, where the most respected manufacturers want to protect their products from counterfeiting. Today there are a number of solutions related to the application of microstructures on the lateral faces of diamonds by which the product can be identified [1], but such marks can be removed quite easily by grinding.

In this work, the characteristics of luminescent defects in artificial diamonds obtained by irradiating the volume of diamonds with femtosecond laser pulses are carried out [2]. In the future the resulting defects can be used to record identification marks.

This work was supported by Russian Science Foundation (grant # 21-79-30063)

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Photoluminescent mapping of optical centers in natural diamond excited by femtosecond laser pulses

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Over the years, many methods have been developed for mapping both natural and synthetic diamonds. However, the most informative methods of gemological analysis of diamonds are optical methods, such as birefringence [1], Raman spectroscopy [2], IR spectroscopy [3], optical absorption [4], and the most sensitive of all the above mentioned methods – photoluminescence (PL) [5]. The classical luminescence method is based on the forced removal of electrons from a stable state to an "excited" state. The relaxation of an "excited" electron proceeds with the release of energy, the spectral range of which mostly corresponds to visible radiation. The process of electron excitation occurs under the influence of various radiation sources (UV lamp, X-ray tube, electron gun, etc.), including those in the visible spectral frequency range such as lasers [6].

In this work, an experimental testing of the method of multidimensional PL in the volume of natural diamond with reference to spatial coordinates was carried out in order to map optical centers [7]. A femtosecond laser was used as a monochromatic source of ultrashort pulses for exciting PL. It generates radiation at two harmonics: $\lambda_1 = 1030$ nm and $\lambda_2 = 515$ nm, respectively (a duration of ~ 300 fs, a repetition rate ~ 100 kHz). PL radiation was transferred from the bulk of natural diamond to the slit of the spectrometer via a UV quartz microscope objective, mounted perpendicular to the axis of propagation of laser radiation. The photoluminescence analysis was performed using an ASP-150F broadband spectrometer. The diamond was scanned in a plane perpendicular to the propagation axis of laser ultrashort pulses and the observation axis.

The PL spectrum was used to analyze natural diamond. The step between the measurement points is ~ 200 µm. Comparing the cross sections of the maxima of the PL spectrum amplitude at each point, one can draw a conclusion about the homogeneity of diamond and the presence of optical centers. Optical centers cause local drops in the global maximum of the PL spectrum amplitude. By the magnitude of the decrease in amplitude, one can judge the characteristics of the optical center, and thanks to linear translators of displacement, one can estimate the size in three coordinates.

This work was supported by Russian Science Foundation 21-79-30063.

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Engineering of atomic defects in the crystal structure of diamond

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The regularities of the transformation of atomic defects in the structure of laboratory-grown and natural diamonds have been studied under two main methods of exposure: radiation damage and high-temperature annealing, which provide almost exhaustive possibilities for controlling defects in diamond. As a result, a material with a given set of properties can be obtained, which is promising for various ultra high-tech applications.

Section 8: Ultrafast laser technologies in biomedicine

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Scope

Ultrafast laser biophotonics

Ultrafast laser fabrication of bionanomaterials

Femtosecond laser biosensing

Ultrafast theranostics

In vivo staining-free visualization of mast cells in human skin using two-photon femtosecond-pulsed tomography with fluorescence lifetime imaging

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Mast cells (MCs) are multifunctional immune cells located in all tissues of the body. In the skin, MCs are located in the dermis in resting and activated states and contribute to the pathology of several diseases including urticaria, psoriasis, atopic dermatitis and mastocytosis where the density of MCs is increased at lesional sites. In the activated state, mast cells release their granular constituents, which triggers inflammatory reactions. Assessment of MC number and their activation state is currently limited to histomorphometric analysis of skin biopsies and *in vivo* visualization in the skin is not applicable.

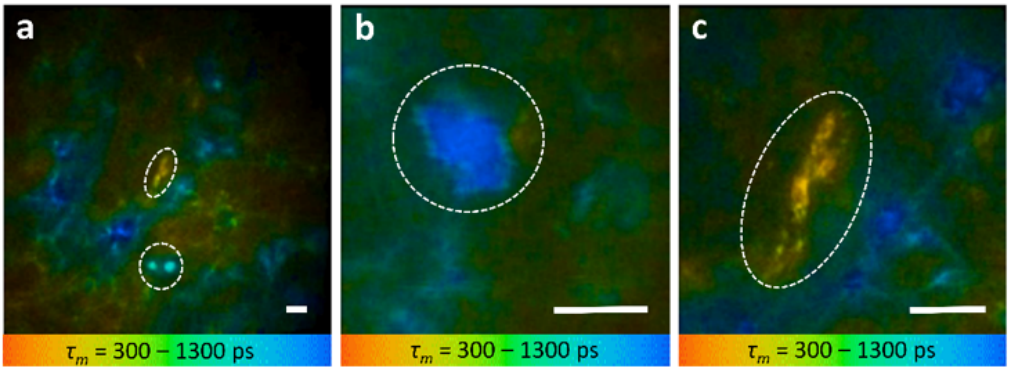

Figure 1: TPE-FLIM images (mean fluorescence lifetimes τ_m in a color gradient from 300 to 1,300 ps) of the human dermis at ≈90 µm measured *ex vivo* without staining on the inner forearm of a 40 y. o. healthy male volunteer. The marked dotted areas show the MCs (a), resting MC (b) and activated MC (c). Scale bar: 10 µm.

Using label-free non-invasive two-photon excited fluorescence lifetime imaging (TPE-FLIM), we have identified mean fluorescence lifetimes of resting (τ_m = 1248±287 ps) and activated (τ_m = 862±268 ps) MCs in cell culture *in vitro* and in skin biopsies *ex vivo*. Then, based on determined TPE-FLIM parameters, we have visualized and quantified MCs and their activation states in the papillary dermis of healthy volunteers, as well as in mastocytosis and allergy patients for the first time, *in vivo* (Fig. 1). The TPE-FLIM imaging was provided using a tunable excitation laser operated at 760 nm and generating 100 fs pulses at a repetition rate of 80 MHz. TPE-FLIM parameters of MCs are distinct and can be differentiated between resting and activated MCs and other dermal cells with a sensitivity of 0.81 and 0.87 and a specificity of 0.85 and 0.84, respectively [1]. The developed method may become an important tool for non-invasive *in vivo* diagnostics and therapy control in dermatology and immunology that target MCs.

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Optical properties of germanium nanocrystalline films and nanoparticles prepared by nanosecond lased deposition in inert gas atmosphere

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Nanostructured semiconductor materials possess a wide range of potential applications. After the experimental observation of visible luminescence from porous Si by Canham [1], investigations of nanostructures of the semiconductor elements of group IV have been actively developed. Recently, we have demonstrated that pulsed laser deposition (PLD) in inert gas atmosphere of low pressure could be used to prepare highly luminescent films of Si nanoparticles (NPs) for potential biomedical applications [2]. However, crystalline germanium (Ge) NPs are studied less than silicon (Si) ones. Bulk c-Ge is an indirect gap (0.67 eV) semiconductor with effective Bohr radius of about 25.3 nm [3] that results in a stronger effect of the quantum confinement compared to c-Si. For biophotonics application it is important that Si and Ge NPs are nontoxic and biodegradable.

In presented paper, Ge NPs were deposited on c-Si substrates by PLD in inert gas atmosphere (He or He+N_2) with different pressure. Then the deposited films were dispersed in ethanol to Ge NPs. The deposited Ge films and NPs were investigated by means of the scanning electron microscopy, X-ray diffraction, Raman and photoluminescent spectroscopy.

The size of Ge NPs was found to be about 50 nm. The Raman spectra of as-deposited Ge films and Ge NPs demonstrated the presence of nanocrystalline form of Ge, that was also confirmed by XRD data. The photoluminescence spectra of the samples consisted of lines in the visible spectral region, which can be associated with optical transitions via defect states in germanium oxide shell of Ge NPs. The obtained results indicate that PLD method is promising to prepare Ge nanostructure for biophotonic and biomedical applications.

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Physical methods against microbial communities in food production

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Every year, disinfection methods are improved and stricter production control programs are introduced, but despite the measures taken, the circulation of pathogenic microorganisms in food enterprises does not decrease.

Researchers around the world in search of the root cause of the constantly occurring infection are increasingly claiming that for most pathogenic microorganisms, the cause is their ability to form stable forms of existence – bacterial biofilms. Unlike planktonic cells, biofilms are resistant to the effects of disinfectants [1,2].

The most promising direction in the fight against biofilms in recent years is the use of physical and chemical methods. Among the physical methods, the use of nanostructured surfaces and nanoparticles of metals and semiconductors with antibacterial properties is a promising direction. The paper presents the results of studies of the antibacterial properties of nanoparticles on food pathogens.

The research on food pathogens with nickel-oxide NPs was supported by the Ministry of Science and Higher Education of the Russian Federation (Project No. 075-15-2020-775).

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How surface relief affects wettability and cell behavior on ultra-short laser-textured surfaces

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Femtosecond lasers are known to be versatile tools capable to form a variety of hierarchical micro and nanostructures on solid surfaces allowing an efficient control over various surface properties. Among these properties, wettability plays a crucial role in the development of numerous application not only in the industry, but also in the biomedical field. Among other factors, wettability strongly affects the behavior of cells, bacteria and even of viruses. A controllable modulation of the wetting properties affects both osseointegration and mechanotransduction, which are known to be key process in dental and orthopedic implant integration, as well as in the bioengineering in general.

Despite a large number of promising experimental results, it is still unclear how to predict and explain the wettability of laser textured surfaces. The classical models, such as the Wenzel and Cassie-Baxter, contain adjustable parameters and provide only rough explanations. The development of more realistic models remain challenging because of the lack of understanding of the effects of both surface and liquid properties on the behavior of a liquid droplet on the surface. In particular, the difficulty arises from the change in surface wettability with time after laser treatment. To better understand this process, we use a continuum-level modeling [1] to study the wetting dynamics of a liquid droplet on laser-textured Ti, Ti alloys, as well as on several other surfaces. The droplet dynamics and the calculated evolutions of the contact angle with time for both structured and non-structured surfaces are examined and are compared to a set of experiments [2]. Our results demonstrate the role of the surface relief and composition. Several explanations are proposed based on the performed modeling, which is shown to be not only suitable for the better understanding of the process involved, but can be also used for the implant relief optimization.

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Laser-induced forward transfer for antibacterial application

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Today society is recognizing antibiotic resistance as a threat to modern medicine. Especially dangerous are those that can form a community called biofilm. Biofilms are highly resistant as of their formation and special tactics to keep community from dying [1]. During the pandemic, many people were prescribed antibiotics for preventative reasons turning out the number of cases with concomitant bacterial infection is less than 10% [2]. This and many other reasons such as lack of antibacterial development, is forcing scientists to look for new ways to fight bacteria.

Bactericidal effect of some metal nanoparticles (such as silver, nickel, and cuprum) is well-known. Laser induced forward transfer (LIFT) method is based on the transfer of metal nanoparticles onto the biofilm under the influence of the focused laser irradiation with definite fluency, frequency, and speed from the "donor" substrate (in our case, in the form of a sputtered thin metal film on the polymer matrix composite) to the substrate where the pattern is going to be formed (acceptor substrate) [3]. In this work LIFT technique study is carried out and the dependence of the radiation power and the thickness of the metal layer was considered.

As a source of radiation 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 was used. The radiation was focused by a galvanoscanner with a lens focal length of 160 mm. Obtained samples were visualized using Live/Dead viability kit.

The research was supported by the Ministry of Science and Higher Education of the Russian Federation (Project No. 075-15-2020-775).

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Molecular dynamic simulation for estimation of porosity effect on laser ablation threshold in silicon

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Laser ablation is one of the rapidly developing and perspective methods for nanofabrication. This method is can be used both for structuring substrates and for the synthesis of nanoparticles with different morphologies. The molecular dynamics simulation models attract an increasing interest since they allow to model many processes, such as melting, evaporation, the appearance, and behavior of phonons, the behavior of dislocations in crystal lattices, as well as to predict the reactivity and properties of compounds using only a few parameters for the description of atoms interaction.

In the present work, the ablation of a porous silicon target under irradiation with ultrashort laser pulse is simulated using the molecular dynamics approach. The number of ablated atoms is calculated for targets with different porosity under irradiation with wavelengths in the ultraviolet (UV) and visible spectral ranges, which correspond to stronger and weaker absorption coefficients, respectively. It is found that an increase of the porosity to 80% leads to a 1.5–3 times decrease of the ablation threshold compared to the bulk silicon, while a decrease of pores size from 2.5 to 1.2 nm leads to the stronger ablation threshold drop and the effect is stronger for the UV irradiation.

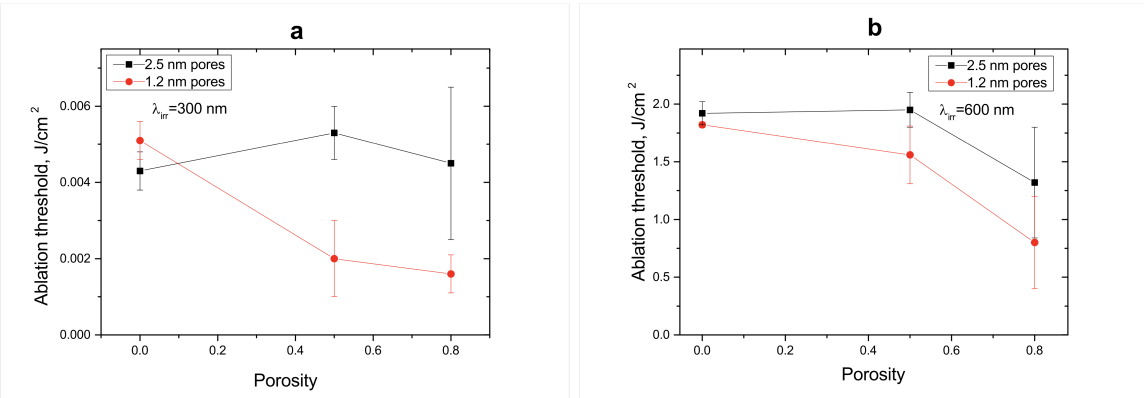

Figure 1: Ablation thresholds for the irradiation with wavelength λ_{irr}=300 nm (a) and λ_{irr}=600 nm (b) vs target's porosity.

Figure 1 shows the dependences of the ablation threshold on porosity for two irradiation wavelengths and two pore sizes. In contrast to the irradiation into the visible range (Fig. 1b), the laser ablation with UV light (Fig. 1a) results in stronger dependence of the ablation threshold on target porosity. The effect is even more pronounced for smaller pore sizes. This ablation threshold decrease is explained by the large contribution of the surface and the weakening of the average binding energy in the substance. Reducing the ablation threshold can be important in the laser ablation synthesis of nanoparticles due to lowering the laser requirements for the ablation.

The reported study was funded by RFBR, project number 20-02-00861.

In vivo visualization of fibroblasts in tattooed human skin using two-photon femtosecond-pulsed tomography with fluorescence lifetime imaging

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Tattoos and body art are increasingly common, yet the influence of tattoos and tattoo inks on the skin is not well established, partly due to missing visualization methods [1]. For the first time, we visualize carbon black tattoo ink nanoparticles in human skin in vivo and non-invasive using fs-pulsed two-photon excited fluorescence (TPE-AF) and fluorescence lifetime imaging (TPE-FLIM). The tunable titanium sapphire laser at 760 nm, generating 100 fs pulses at 80 MHz (Fig. 1a) provides an imaging depth of ≈150 µm in the skin [2]. TPE-AF and the fluorescence decay analysis of single fs-pulses in TPE-FLIM are specific to visualize dermal cells and components and are superior in spatial resolution to single-photon excited microscopy [3].

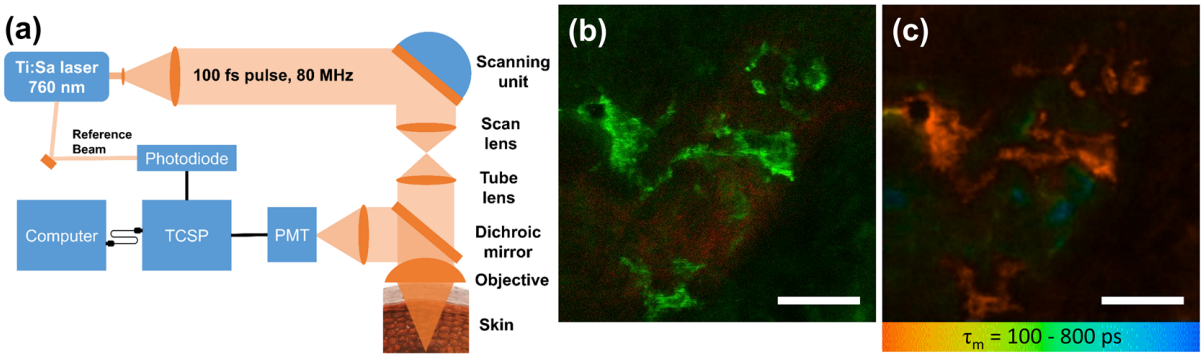

Figure 1: Schematic illustration of the TPE-FLIM tomograph (a). TPE-AF (b) and TPE-FLIM (c) images of carbon black nanoparticles-loaded fibroblasts (green in (b) and red in (c)), located between collagen I matrix (red in (b)) measured *in vivo* in a 24 months old tattoo. Scale bar 12 µm.

Both, the cellular intake of carbon black nanoparticles and the structural changes of dermal collagen I are categorized with intravital imaging. Carbon black nanoparticle agglomerates were successfully identified in the fibroblasts, making them visible by TPE-AF (Fig. 1b) and TPE-FLIM (Fig. 1c). Carbon black was additionally identified inside the keratinocytes and Langerhans cells in the epidermis and inside the mast cells and macrophages in the dermis. In native skin, dermal cells were classified by their morphology and distinct TPE-FLIM parameters [3]. However, in tattooed skin, cells are able to absorb/phagocytose tattoo inks and keep them for their lifespan, which was measured by the short TPE-FLIM and enhanced TPE-AF intensity. The extracellular matrix was found to be free of tattoo inks after the initial skin regeneration of 3 weeks. The TPE-FLIM technique could be useful in clinical dermatological practice for managing patients with tattoo-related complications and to give useful information about cellular pathways of exogenous substances in the skin, their accumulation and understanding the skin regeneration after trauma.

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Neuroport with fibre-optic internal structure for therapy and prevention of brain tumours

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The main difficulty in treating brain tumours is their active invasion and the high risk of metastasis. In turn, their invasion into the deep tissue often occurs via channels in the white matter tissue of the brain, which is similar in structure to blood vessels.

Thus, the spread of cancer cells deep into the tissue occurs through structures that have a tubular structure, suggesting that directed cell growth along such structures is possible. The approach of using optical fibres as guide rails for cancer cell growth is proposed and tested in this work. In this way, it is possible to direct the growth of cancer cells from deep in the tissue to the outside, and thus to carry out prevention and therapy of brain tumours. In turn, optical fibres are able to deliver drug and laser radiation and thus enable photodynamic therapy and diagnosis of the tumour bed.

Thus, the multifunctionality of optical fibres has been investigated in work on experimental animals as well as on cell cultures. In addition, the interaction between biological structures and fiber structure was studied by evaluating the dynamics of fluorescence signal kinetics of photosensitizer, namely, fluorescence lifetime of different cellular structures in different environment. Thus, the work shows the possibility of creating multifunctional implants for the therapy and prevention of brain tumours with the tracking of functional changes in tissues by estimating the lifetime of the photosensitizer [1].

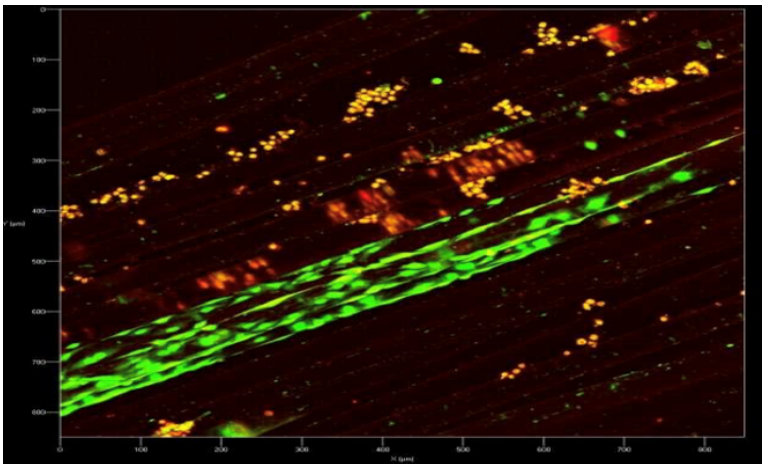

Figure 1: Optical fiber neurosystem for deep-lying brain tumors phototheranostics.

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Metal nanoparticles for antibacterial treatment

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Today the increasing resistivity of different bacterial strains against various antibiotics become a large problem [1]. Bacteria are especially dangerous in biofilms due to the increased resistance to methods of combating them [2]. Are known, that many metal nanoparticles have antibacterial properties. One of the mechanisms responsible for the death of bacteria is the generation of singlet oxygen by nanoparticles.

In this work silver and copper nanoparticles were produced by method of laser ablation in liquid and through the method of laser-induced forward transfer. Nanosecond and femtosecond laser were used as a source of laser radiation. Studies were carried out on gram- positive and gram- negative bacteria.

The research was supported by the Ministry of Science and Higher Education of the Russian Federation (Project No. 075-15-2020-775).

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Structure images analysis of collagen using two-photon microscopy and machine learning approaches

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This work demonstrates the results of comparing images for healthy and lymphedematous skin of rats obtained by a two-photon microscope. Our study was carried out on 16 outbred sexually mature white rats of both sexes weighing 200–250 g. The rats were divided into 3 groups: 6 control rats, 6 rats with a model of hind limb lymphedema with radiation therapy, and 4 rats with a model of hind limb lymphedema without radiation therapy. Skin measurements at several depths (30, 40, 50 µm), 5 stacks for each rat, were performed using a two-photon microscope MTPflex (Jenlab Germany) [1]. The difference in the images was analyzed using gradient methods: convolution with the Sobel operator, the Canny boundary detector, the method of oriented gradient histograms [2]. There are first-order statistics and support vector machines used to compare the results of applying gradient methods. When comparing, the data were divided into 3 groups, a group for training (30% of the data), validation (20% of the data), and a group for testing (50% of the data). As a result, it was shown that collagen destructuring occurs in lymphedema, which can be numerically noted using gradient methods.

The research was carried out with the support of a grant under the Decree of the Government of the Russian Federation No. 220 of 09 April 2010 (Agreement No. 075-15-2021-615 of 04 June 2021).

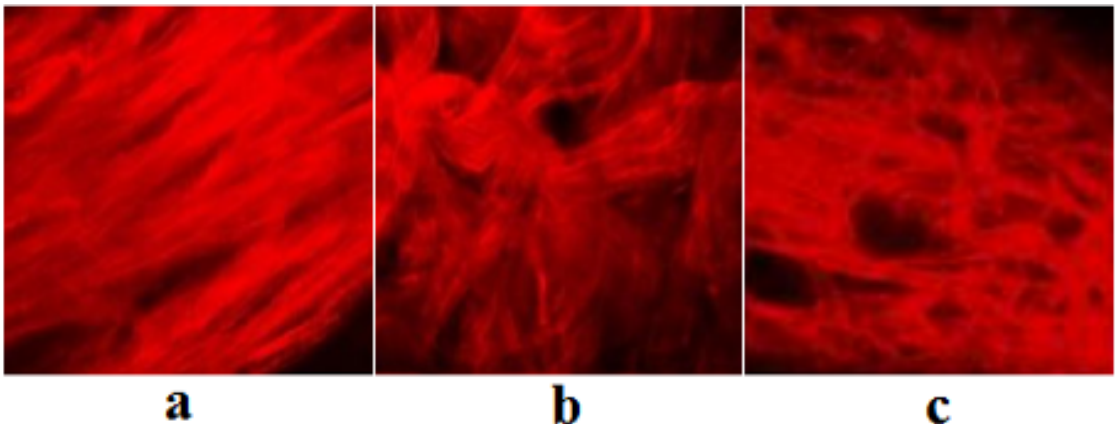

Figure 1: The example of images of healthy tissue (a) tissue after removal of lymph nodes without irradiation (b), after removal of lymph nodes and after irradiation (c).

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Investigating monocyte metabolic state using fluorescence lifetime unmixing

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Cofactors like NADH and flavins are known to be an indicator of the metabolic state of cells. Their fluorescence lifetime greatly depends on their binding to proteins. Glycolysis results in a high ratio of unbound NADH to bound one, while for the oxidative phosphorylation the ratio is opposite. The fluorescence lifetime of unbound NADH is about 0.4 ns, while for bound NADH it is about 1.2 ns.

Currently existing methods utilize fluorescence decay fitting with two exponents and calculate a ratio between the amplitudes for the short and the long lifetime kinetics. These methods require the presence of both NADH fluorescence decays and the absence of other sources of fluorescence.

We propose a method of fluorescence lifetime decomposition based on the Maximum Entropy Method (MEM) [1]. MEM seeks to find the distribution of lifetime amplitudes that fits the data while maximizing its information entropy. The resulting distribution doesn't require prior knowledge on the number of fluorescence lifetime components.

NADH fluorescence lifetime images of THP-1 monocytes were obtained using LSM-710 laser scanning microscope (Carl Zeiss) with FLIM attachment (Becker-Hickl) under femtosecond laser excitation (Chameleon Ultra II, Coherent). The images were processed using MEM (Fig. 1). This allowed separating the signals of bound and unbound NADH.

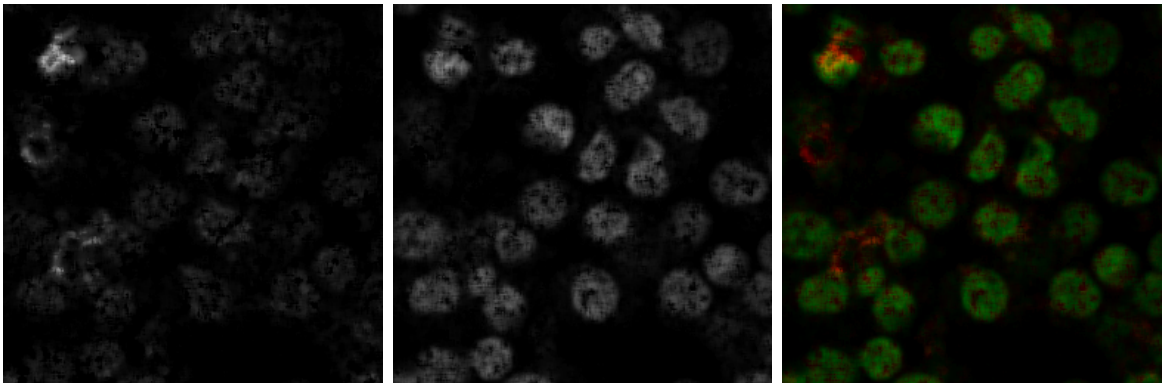

Figure 1: THP-1 monocyte cells NADH fluorescence lifetime unmixed image: unbound NADH (0.2–0.8 ns range, left), bound NADH (0.9–2.0 ns range, center). Combined image (right) displays unbound NADH in red, bound—in green.

This work was supported by RFBR, № 20-02-00928.

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Laser-based inactivation method of pathogenic microorganisms

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Searching and developing methods for combating pathogenic microorganisms remain relevant accordingly the emergence new and mutations of existing types of bacteria and viruses. Well-studied and widely used method of UV disinfection has the characteristic disadvantages: UV affects both pathogenic and mammalian cells, and it is the main cause of cancer development. In addition, it is known, that there are many types of bacteria and viruses are resistant to UV [1]. Method of selective inactivation of pathogenic microorganisms by ultrashort laser pulses (USP) of the visible and near IR regions has been already developed. It is based on excitation of mechanical vibrations in protein molecules as the result of the effect of impulsive stimulated Raman scattering (ISRS). This method is safe for mammalian cells, but it has complex and expensive equipment and long time of exposition ($\sim$ 1 hour) [2].

In this work, we investigated the possibility of inactivation of pathogenic bacteria by exposing it to femtosecond laser pulses of mid-IR range a low average power and wavelength of 6 µm. Mechanism of inactivation is the selective denaturation of functional proteins of bacteria as a result of the destruction of hydrogen bonds, which responsible for the secondary and tertiary structure [3]. Comparison of the stationary and dynamical transmittance spectra of bacterial layer for various values of peak intensity USP was performed. Exposition of pulses a low peak intensity (0.3 GW/cm^2) led to an increase in characteristic absorption in the range 1550–1900 cm^{-1}. Blue shift, which related to the destruction of hydrogen bonds, has been observed.) Strong modification upon the exposition of high intensity pulses (5 GW/cm^2) was observed. It has a reversible character. Results of microbiological investigated showed a decrease of the number of colony-forming units depending on peak intensity of USP.

The research was supported by the Ministry of Science and Higher Education of the Russian Federation (Project No. 075-15-2020-775).

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Numerical simulation of photothermal effect in presence of silicon nanoparticles in skin tumors

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Photothermotherapy (PTT) techniques are based on using inorganic nanomaterials as a thermal coupling agents for a biological sample [1]. Silicon is material of high biocompatibility, bioavailability, biodegradability and low toxicity [2]. In this study nodular basal cell carcinoma (BCC) laser heating in presence of silicon nanoparticles is simulated for temperatures corresponding to mild hyperthermia regimes (42–43°C). The study was performed for nanoparticles obtained by picosecond laser ablation of silicon nanowires in water and ethanol. The optical properties of suspensions were obtained through Mie calculations based on the atomic force microscopy data on particles size distribution.

We used Monte Carlo simulation method to obtain volumetric density of absorbed energy distribution as a result of light absorption from continuous laser source. Then we solved time-dependent bioheat equation through COMSOL® Multiphysics package. Simulations revealed that irradiation by moderate laser powers of human skin containing carcinoma with size of several millimeters in presence of silicon nanoparticles formed by pulsed laser ablation of low-doped silicon nanowires in water in tumor tissue allows to reach the temperature of 42°C in all tumor volume, while without nanoparticles at the same irradiation conditions the full heating of considered carcinoma is not possible. The optimal laser powers were 100 mW for beam of 5.1×5.1 mm^2 and 170 mW for beam size of 10 mm^2, and the irradiation time was 600 sec. At these power values the maximal increase in heating efficiency of tumor tissue was 1.5°C and 1°C, respectively.

This work was supported by Russian Science Foundation (grant № 19-12-00192).

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Comparative FT-IR spectroscopy of Gram-positive and Gram-negative bacteria after laser-induced forward transfer of Cu and Ag nanoparticles

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Biofilms form when planktonic bacteria find optimal conditions for procreation and generate a durable extracellular matrix, consisting of polysaccharides, proteins and parts of DNA [1,2]. Fourier infrared (IR) spectroscopy is a great tool for identification of a great number of chemical compounds, bacteria, yeasts etc. [3]. It can also demonstrate the difference between alive and dead bacteria, which is indicated by the shift or lower intensity/complete disappearance of several spectral peaks. So, we have produced the comparative Fourier-transform IR spectroscopy of several Gram-positive (*Staphylococcus aureus, Listeria monocytogenes*) and Gram-negative (*Pseudomonas aeruginosa, Escherichia coli, Salmonella typhimurium*) bacterial biofilms before and after treatment with Cu and Ag nanoparticles (NPs), applied to biofilms by laser-induced forward transfer (LIFT). Analysis of FTIR spectra showed the rupture of the cell membrane, accompanied by the emerging of peak, corresponding to amide I molecular oscillations, which may have resulted in bacterial death. Each sample was characterized by scanning electron microscopy before and after NP treatment.

The research was supported by the Ministry of Science and Higher Education of the Russian Federation (Project No. 075-15-2020-775).

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Search for alternative methods of controlling biofilms of pathogenic microorganisms

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Antimicrobial resistance is one of the major global health security risks. Especially resistant to the action of antibacterial drugs are pathogens capable of forming bacterial communities or biofilms. The role of infections caused by pathogens existing in biofilms is very large [1]. One of the possible alternatives to the use of antibiotics is the use of metal and semiconductors nanoparticles. In our studies, we have shown high antibacterial and antibiotic film activity of silver, copper, zinc and silicon nanoparticles.

The method was successfully tested on almost all microorganisms belonging to the ESCAPE group: *Enterococcus faecium, Staphylococcus aureus, Klebsiella pneumoniae, Acinetobacter baumannii, Pseudomonas aeruginosa* and *Enterobacter spp.* The transfer of nanoparticles by the above-mentioned lone method is effective both for planktonic bacteria and biofilms.

The research on the LIFT technology and through-flow sterilizer was supported by the Russian Science Foundation (grant № 18-15-00220), on food pathogens with nickel-oxide NPs – by the Ministry of Science and Higher Education of the Russian Federation (Project No. 075-15-2020-775).

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Stimulated low-frequency Raman scattering in biological nanoparticles

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Biological nanoscale and submicron particles (viruses, proteins, DNA and RNA molecules, cellular vesicles, ect.) are the objects of intensive theoretical and experimental research primarily because of their role in everyday human life. One of the most important tasks in this research is the development of new methods for biological nanosized objects characterization, identification and impact on its. Numerous methods of linear and nonlinear optics are very effective for these proposes.

Low-frequency Raman scattering (LFRS) [1] can be used as effective tool for investigation various types of nanometer (1–100 nm) and submicron (100–1000 nm) sized systems. LFRS is a result of inelastic light scattering by acoustic vibrations of nanoparticles. Its frequency shift corresponds to the eigenfrequencies of acoustic vibrations of nanoparticles, which are in the terahertz or gigahertz frequency range. Since acoustic eigenfrequency is determined by the morphology of the objects under study, such important characteristics as the size and sound velocity can be determined from the LFRS spectra. As it was shown for nanoscale biological objects both the frequency and damping of the vibrational modes are significantly affected by the properties of the surrounding medium. In this work we report on application stimulated low frequency Raman scattering (SLFRS) [2] which is a stimulated analog of LFRS for the use both for the tasks of determining the elastic characteristics of biological systems in order to identify them, and for the tasks associated with the creation of a biharmonic pumping source for the purpose of resonant and selective impact on such systems.

In this study SLFRS characteristics in the suspension of various types of plant viruses and proteins were experimentally investigated. The SLFRS frequency shifts, corresponding to acoustic eigenfrequencies of the samples, conversion efficiency and thresholds of excitation were measured. The experimental parameters are in good agreement with the values calculated using the Lamb theory [3], taking into account the influence on the elastic characteristics of biological particles of the environment.

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Ultrafast energy transfer determines the formation of fluorescence in natural organic matter

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Humification is a ubiquitous natural process of biomass degradation that creates multicomponent systems of natural organic matter (NOM) in water environments, soils, and organic rocks. Despite significant differences in molecular composition, the optical properties of NOM of different origin are remarkably similar, and the reason for this remains largely unknown. Here, we employed fluorescence spectroscopy with (sub)picosecond resolution to elucidate the role of electronic interactions within NOM. We revealed an ultrafast decay component with a characteristic decay lifetime of 0.5–1.5 ps and spectral diffusion originating from excitation energy transfer (EET) in the system. The rate of EET was positively correlated to the fraction of aromatic species and tightness of aromatic species packing. Diminishing the number of EET donor–acceptor pairs by reduction with NaBH4 (decrease of the acceptor number), decrease of pH (decrease of the electron-donating ability), or decrease of the average particle size by filtration (less donor–acceptor pairs within a particle) resulted in a lower impact of the ultrafast component on fluorescence decay. Our results uncover the role of electronic coupling among fluorophores in the formation of NOM optical properties and provide a framework for studying photophysical processes in heterogeneous systems of natural fluorophores.

This work was supported by the Young Investigator Research Grant 2017 from the International Humic Substances Society.

Microbiome of food production and innovative methods of microbiological risk management

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The safety of food systems and their resistance to such challenges as outbreaks of infectious diseases for our country, which has enormous natural resources and is actively developing the national agro-industrial complex, is of particular importance. The main task of the food industry is to ensure the microbiological safety of manufactured products, based on the prevention of possible risks of ingestion of microorganisms that cause foodborne infections [1].

The microbial community of objects of food production is characterized. The study of the microflora of food production facilities was carried out on the example of 4 enterprises of the meat and poultry processing industry. More than 80 washes were selected and investigated, 250 colonies were studied and 140 strains of various microorganisms were identified using a set of methods. So the highest prevalence was shown only by 6 groups of bacteria, including: Pseudomonas, Acinetobacter, Enterobacteriaceae, spore-forming bacteria, Staphylococcus spp. and LAB. Less than 10% of the total number of bacteria identified at one processing plant were the following groups of bacteria: *Aeromonas* spp. *Brochothrix* spp., *Microbacterium* spp., *Micrococcus* spp., *Neisseriaceae*, *Psychrobacter* spp., *Ralstonia* spp.,*Rhodococcus* spp., *Shewanella* spp., *Sphingomonas* spp., *Stenotrophomonas* spp. and *Vibrio* spp. [2].

A complex of studies was carried out to study the antimicrobial properties of various materials, including polycationic polymers and nanoparticles, synthesized as a result of the use of physical methods, as well as to study the antimicrobial effect of plasma discharge and ultrasound in a liquid. Using polycationic polymers as an example, a comprehensive approach to assessing antimicrobial activity using microbiological methods, flow cytometry, and fluorescence microscopy has been tested. The antimicrobial activity of polymers on planktonic forms of microorganisms (*L. monocytogenes* and *P. aeruginosa*) isolated from objects of the production environment, as well as on the formed and emerging biofilms of these microorganisms, was studied. Revealed a higher antimicrobial activity of polymers in relation to gram-negative microorganisms.

The research on food pathogens with nickel-oxide NPs was supported by the Ministry of Science and Higher Education of the Russian Federation (Project No. 075-15-2020-775).

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Fabricating nanoparticles for biophotonics by laser ablation and fragmentation of nano- and microstructured silicon in liquids

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Pulsed laser ablation in liquids (PLAL) is a powerful tool to produce a variety of nanoparticles with desirable size, physical and chemical properties [1]. Silicon nanoparticles (Si-NPs) fabricated by this technique have potential in different biomedical applications [2–4] due to high biocompatibility and biodegradability of nanostructured silicon. Note that high-yield production of Si-NPs is quite time-consuming and requires employment of a powerful laser with high pulse repetition rate. To enhance the efficiency of Si-NPs fabrication, we suggest using preliminary nano- or microstructured silicon instead of traditionally used crystalline silicon targets.

In our work we use porous silicon films, silicon nanowires arrays and mechanically grinded silicon microparticles (1—6 µm in size) as targets for picosecond (1064 nm, 34 ps, 10Hz) and femtosecond (1250 nm, 160 fs, 10Hz) PLAL. Ablation thresholds for the porous silicon and silicon nanowires targets in water and ethanol were demonstrated to be several times less in comparison to those for crystalline silicon, thus providing high-yield Si-NPs production. Similar tendency is observed for laser fragmentation of silicon microparticles in these liquids.

According to scanning electron microscopy and dynamic light scattering studies, the mean sizes of Si-NPs are in the range of 24—340 nm depending on the used target, buffer liquid, laser pulse duration and irradiation time. Raman spectroscopy data analysis revealed almost perfect crystallinity of the formed Si-NPs for silicon nanowires PLAL and laser fragmentation of the silicon microparticles.

The studied Si-NPs exhibit fluorescence emission in the range of 600–900 nm which is most likely caused by internal structural defects in them [3]. Spectrophotometry measurements of the Si-NPs suspensions revealed their effective scattering in the spectral range of 400–1000 nm, which is explained in the frames of the Mie theory. Using the defined scattering and absorption parameters of the Si-NPs suspensions, the heating of tumor tissue with embedded nanoparticles was numerically modelled [4] in order to evaluate their potential in tumor hyperthermia. The obtained results allow to conclude that the Si-NPs fabricated via PLAL are promising either in fluorescence and scattering bioimaging or in photohyperthermia of living organisms.

This work was supported by Russian Science Foundation (grant № 19-12-00192).

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Editors:

Stanislav Bezhanov

Daria Mokrousova

Printing design:

Daria Mokrousova

V International Conference of Ultrafast Optical Science (UltrafastLight 2021),
M. LCC "SAM Polygraphist", 2021. 268p.
ISBN 978-5-00166-456-7.

V International Conference on Ultrafast Optical Science
«UltrafastLight-2021»
Moscow, 2021