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Sections:
1. Ultrafast high-field science
2. Ultrafast phenomena in condensed matter and ionized gases
3. Ultrafast laser nanofabrication and nanophotonics
4. Femtosecond non-linear optics. Filamentation
5. Femtosecond radiation in spectroscopy and optical frequency metrology
International Conference on Ultrafast Optical Science

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1. Ultrafast high-field science section

Chair Valery Yu. Bychenkov

LASER WAKEFIELD ACCELERATION OF ELECTRONS FOR RADIATION GENERATION

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Experiments using the Hercules laser at the University of Michigan have shown that control of the electron beam from Laser Wakefield Acceleration can be used to optimize the generation of betatron x-ray radiation. This can be achieved by: i) extending the interaction length to increase the overall flux, ii) use of ionization injection to produce more x-ray photons, iii) off-axis injection of electrons in a density down ramp to produce a very-monoenergetic and divergent "ring" of high energy electrons which increases the flux of x-rays by about an order of magnitude. We perform experiments to record absorption and phase contrast images of biological samples in particular highlighting the contrast enhancement achievable with the simple propagation technique of phase contrast imaging. Preliminary measurements of time resolved x-ray absorption spectroscopy near the K-edge of a laser heated aluminum plasma using betatron radiation will also be presented.

This work is supported by U.S. Department of Energy/National Nuclear Security Administration Grant No. DENA0002372.
Understanding the collective response of plasma particles to intense laser radiation is both of fundamental interest and important to the development of laser-driven accelerators. The case of ultra-thin foils which become relativistically transparent during the interaction with an intense laser pulse is of particular interest due to their importance in laser-driven ion acceleration.

We report on experimental and 3D particle-in-cell simulation results on the collective motion of plasma electrons and ions responding to the propagation of an intense laser pulse through an expanding ultrathin foil which becomes relativistically transparent [1-2]. It is shown that spatial structure within the beam of energetic electrons produced can be controlled by variation of the laser pulse parameters. Diffraction of intense laser light propagating through a self-generated ‘relativistic plasma aperture’ produces a structured near-field diffraction pattern, to which the electrons collectively respond. Static and rotating electron beam profiles can be induced by variation of the degree of ellipticity of the laser polarisation [1]. The resulting modulation of the charge-separation-induced electrostatic field means that the ion motion can also be manipulated [2]. These concepts have been verified via experimental and 3D particle-in-cell simulations and provide a new avenue for optical control of laser-accelerated electron and ion beams. It is also shown numerically that the spatial, temporal and polarization properties of the transmitted laser pulse are potentially controllable by these relativistic optical and photonics processes.


LASER-PLASMA SOURCES OF HIGH ENERGY ELECTRONS

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The acceleration of electrons to high energies up to the TeV range, as well as the creation of compact sources of relativistic electrons and hard X-ray radiation, requires the development of advanced acceleration methods with a large acceleration gradient much larger than that achievable in conventional radio frequency accelerators. Among these methods, the most actively developing approaches are based on the use of wake fields generated in plasma by relativistic-intense femtosecond laser pulses. In view of current and future experiments, various methods of electrons acceleration in a plasma are discussed. The laser wakefield acceleration of short electron bunches to multi-GeV energies with small emittance and energy spread is modelled and analyzed. Trapping and acceleration of short electron bunches externally injected into the wakefields generated by intense femtosecond laser pulse in plasma channel are optimized. The influence of the laser nonlinear dynamics and loading effect (self-action of the bunch charge) to the bunch final energy and energy spread of the accelerated electrons is investigated. The limitations to the charge of accelerated electron bunch determined by the demand of its small energy spread is found [1].

The recent results on different mechanisms of high-energy electrons generation required for many applications are also discussed.

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NANOSCALE STRUCTURED TARGETS FOR ENHANCED RELATIVISTIC LASER-
PLASMA GAMMA AND HARD X-RAY SOURCE

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The laser-driven plasma gamma and X-ray sources are of wide interest within the last decade. Small size and high spatial coherence of the source, ultra-short duration of the X-ray burst and high brilliance in a wide energy range allow to efficiently utilize the laser-plasma for X-ray imaging with high spatial resolution, gamma radiography, photonuclear reactions excitation etc. However, the lack of luminosity compared to conventional sources (synchrotrons, accelerators etc.) hampers the wide use of laser-based source in varied applications, and today many efforts are made toward the amplification of laser energy conversion into plasma emission.

In our work we experimentally demonstrate the huge enhancement of gamma and hard X-ray yield from plasma, produced by a relativistically intense femtosecond laser radiation onto the surface of nano-modified targets.

The modified targets were prepared via laser ablation, electrochemical etching or deposition of material onto the surface of solids (Si or metals) forming varied types of structures: nanospheres, nanopores, nanograss, foam etc. The main advantages of these techniques is the ability to create large area sample with very high level of structures reproducibility providing high stability of plasma parameters from shot to shot at high repetition rate of pulses.

In our experiments we used a laser pulse delivered by 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 5x10¹⁸ W/cm², contrast on ps timescale >10⁹). Measuring the gamma spectra of laser-produced plasma we found, that the hot electron temperature raised from 200 keV (for initially flat bulk target) up to >600 keV (at irradiation of nanograss target), leading to a substantial growth of
gamma emission (up to tenfold). Such a huge increase is one of the first to be experimentally demonstrated. The highest conversion efficiency into the gamma range (>500 keV) was estimated on the level of 10-5%. At the same time, the hot electrons acceleration efficiency is found to be strongly dependent on type of used nanostructures (nanograss, pores or other).

The experimental results are supported by PIC modeling of intense laser action onto nanostructures, showing the significant increase of hot electron production and energy gain due to efficient acceleration of the particles along the structures by the penetrated field of the laser pulse.

We also demonstrated that the source, formed onto the surface of solids may be utilized for varied applications, including phase contrast image formation. The contrast level of ~20% was achieved when exposing of simple objects to X-ray photons. The gamma photons may be efficiently utilized for radiography of high density objects and low energy threshold nuclear reactions excitations.

The work was supported by RFBR grant 15-02-08113 and Postgraduate Presidential Fellowship for K.A. Ivanov.

**IMPACT OF RADIATION REACTION ON PLASMA WAVES GENERATION BY ULTRA-INTENSE LASER PULSES**

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One of perhaps the most spectacular applications of the novel high power laser facilities is particle acceleration via the longitudinal electric fields generated during nonlinear propagation of intense laser pulses through plasma. But when Lorentz factor of electron quiver motion becomes of the order of few hundreds, radiation reaction effects should also come into play. We study the impact of radiation reaction on propagation of a high intensity electromagnetic pulse through the cold plasma and identify a novel mechanism of longitudinal plasma field generation based entirely on radiation reaction. The regime for which this mechanism dominates over the conventional ponderomotive one is clearly revealed by numerical simulations. We also present a simple analytical model, which can be used to optimize the conditions for plasma wave generation and electron acceleration in a regime when radiation reaction becomes essential.
Proton and ion beams accelerated in hot electron sheaths created on solid targets by ultra-strong laser pulses have remarkable characteristics that have enabled unique applications like ultrafast radiography. The current challenge is to increase the ion energy to and beyond 100 MeV, which is commonly thought to be possible by raising the on-target laser intensity. Here, we are going to present numerical results demonstrating that magnetostatic fields self-generated on a rear target surface may pose a fundamental limit to target normal sheath ion acceleration for high enough laser intensities. Those fields can be stronger than $10^9$ G at laser intensities above $10^{21}$ W/cm$^2$ efficiently magnetizing the sheath electrons and deflecting the protons off the accelerating region, hence degrading the energy transfer from the electrons to the protons. This detrimental effect of the self-generated magnetic fields was first observed in experiments with tightly focused sub-ps laser pulses and then investigated both analytically and numerically [1]. Analysis show that with increasing laser intensity the toroidal magnetic fields generated on the rear surface of the target by hot electron bunches ejected by the laser increase as well but in much faster manner. At $10^{21}$ W/cm$^2$ intensity level they reach Gigagauss magnitudes and begin to significantly affect the electrons trajectories. If the laser pulse is long enough the Larmour radius of the electrons may become comparable to the size of the sheath so that the electrons start to drift and their effective velocities decreases. When the drift velocity become less than the velocity of the accelerated ions the ejected electrons can't overcome ion front and contribute to their acceleration. This is the main mechanism behind the detrimental effect magnetic field has on ion energies. Another mechanism is the deflection of protons away from the central axis where both the electron density and, consequently, accelerating fields are the highest.

In our talk we will discuss how the detrimental effect depends on the main parameters of the laser: intensity, duration and spot width.

The short-pulse lasers that have come in operation recently have provided a new impact to the interaction experiments with targets on a new basis. They require and consume a variety of targets (conventional and original) which provide an interesting data if the targets’ parameters match the laser parameters sharply. Some most fruitful target decisions are plastic aerogels, nanostructured and complex layered structures. These targets are overviewed, the target community for the special targetry works is described, and inspiring selected and expected laser experiments are presented.

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DIRECT ELECTRON ACCELERATION FOR DIAGNOSTIC OF LASER PULSE CHARACTERISTICS

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Contemporary laser facilities allow achieving high spatial-temporal energy concentration. In the same time generated intensive laser pulses can’t be measured by traditional techniques, this fact causes new methods to be of high demand. In the present work we propose a new method of laser pulse diagnostics via direct electron acceleration. Dynamics of electrons accelerated from ultrathin nanofoil are described by the test particle method that is possible when plasma field can be neglected as a comparison with the force of laser-particle interactions. Thereat electron characteristics are determined by parameters of the laser field that is important for the laser pulse diagnostics. Sometimes simulations of the focused laser pulse are based on the paraxial approximation, but the regime of the most tightness focusing on the diffraction limit stays out of its applicability range. To make our model match the experiments we use Stratton-Chu integrals for the description of the laser pulse focused by off-axis parabolic mirror. It allows us to simulate laser-particle interactions with different pulse parameters including its spatial-temporal profiles. Varying laser characteristics it has been shown which of them could be determined at the same time.

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EFFECTS OF IONIZATION AND DENSITY SCALE LENGTH ON ION ACCELERATION FROM LOW-DENSITY TARGET

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In this talk we present results of 3D PIC simulations related to a new mechanism of synchronized ion acceleration be slow light from low-density targets (SASL). We show that account of target ionization as well as change of hydrogen concentration inside target has little effect on maximum proton energy accrued during acceleration in SASL regime. At the same time even small density gradient at the target front may result in significant reduction of maximum proton energy in compare with a case of ideal target shape.

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GENERATION OF SHORT ELECTRON BUNCHES BY A LASER PULSE CROSSING A SHARP BOUNDARY OF PLASMA

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The interaction of a packet of strong electromagnetic waves with a semibounded plasma, which is characterized by a sharp up-ramp density gradient, was studied. The results of the investigation of the process of the generation of a short electron bunch by a laser pulse of relativistic intensity that passes through a sharp boundary of plasma revealed some principal features of the physical mechanism that underlies this process [1]. It has been demonstrated that the process of the self-injection of an electron into the wake wave is fully determined by the characteristics of the plasma oscillators that are excited by the laser pulse that passes through a semi-bounded plasma. A necessary condition for the self-injection of electrons in the first period of the wake field is that the energy of plasma oscillators should exceed the gamma-factor of the wake wave of the laser pulse. It has been established that the process of electron self-injection into the wake wave starts with an electron that initially occurred in the depth of the plasma at a distance from its boundary equal to the amplitude of its subsequent oscillations caused by the interaction with the laser pulse. In what follows, this electron becomes a leader, that is, the first particle in the head part of the bunch trapped in the wake wave. The major fraction of electrons trapped in the wake wave are electrons that initially (in the plasma that is not disturbed by the laser pulse) occurred in front of the electron leader; their order in the trapped bunch is opposite to that in the undisturbed plasma. The length of the trapped electron bunch is determined by the effect of kinematic grouping, which consists in the fact that electron self-injection into the wake wave occurs at the point of space and the moment of time when the electron-leader along with the previously trapped electrons is close to this point. Subsequently, during acceleration of trapped electrons in the wake wave, the length of the bunch increases as a result of the initial spread in the conditions of electron injection and the mutual repulsion of electrons in the bunch, but tends asymptotically to a certain limit. The asymptotic value of the bunch length is determined by the characteristics of the oscillator excited by a laser pulse in a plasma. The spread of energy between electrons in the bunch due to their repulsion increases monotonically during their acceleration in the wake wave in proportion to the length of acceleration. However, the relative electron energy spread in the bunch over large acceleration lengths can be minimized due to analogous growth in the average energy of electrons in the bunch.

Estimations show that a laser pulse that interacts with semi-bounded plasma according to the mechanism that was considered can generate electron bunches with a duration below 1 fs and a charge of several hundred pico-Coulombs.

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Study of radiation properties of solid dense plasma irradiated by ultraintense lasers has a great interest both from fundamental physics and different application point of views. The recently upgraded a petawatt-class J-KAREN-P laser could produce femtosecond pulses reaching the intensity of $10^{22}$ W/cm$^2$ within a micron focal spot and a laser pulse duration of 35 fs. Used high-resolution X-ray spectroscopic methods allow to investigate the ionization properties of medium (Al) and high z (Ti, Fe, Ag, Au) thin foils, identify some parameters of generated plasma from front and rear sides of targets. Provided experiments demonstrated the availability of the laser prepulses drastically decrease the degree of plasma ionization. It was also found that even rather small displacement of laser focus spot from the optimal one strongly reduce intensity of X-ray radiation and plasma ionization. 2D PIC code simulations of femtosecond laser interaction with various materials were provided and compared with experimental results.
ELECTRONS ACCELERATION AND OPTICAL HARMONICS GENERATION IN THE RELATIVISTIC LASER-PLASMA OF SOLID TARGETS

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1. Introduction

The electron bunches with high energy and charge may be generated in low dense plasma via different mechanisms of laser acceleration. In this paper we present the numerical simulations of the possibility of generation MeV electrons at an intensity around 10^{18} W/cm² in an near-critical long pre-plasma.

2. Simulation parameters

We made interaction simulations using the fully relativistic 3D3V PIC code “Mandor,” reduced to the 2D3V within most of our calculations. The simulation box size was 31 mkm (x) × 14mkm (y), spatial resolution was λ/100 and total number of particles was 10^8. Temporal resolution was 3×10⁻³ fs. The planar foil target consisting of cold ions and electrons. A p-polarized laser pulse with duration of 50 fs (FWHM) and central wavelength of 1 mkm entered the simulation box. Varying the laser pulse intensity (10^{17}-10^{19} W/cm²), the initial electron density scale length (0.5-10 λ), incident angle (150-750) and beam waist diameter (2-10 λ) we found the dependences of the hot electrons and optical harmonics yield on these parameters.

3. Results

Analysis of the electromagnetic fields and electron density spatial and temporal distributions from simulation revealed that hot electrons are generated due to the breaking of plasma waves. At the plasma scale length ~1-5λ there are two mechanisms of plasma wave excitation in obliquely incident interaction, appropriate for hot electrons generation: two plasmon decay instability (TPD) and charge separation near the turning surface. Both processes lead to generation of the optical harmonics: 3ω/2 and ω/2 for the scattering of the fundamental on TPD waves and 2ω for the turning surface waves. Also it is shown that electrons with maximum energy are accelerated in crossed fields of incident and reflected laser pulses after wavebreaking, and there is an optimum beam waist diameter for this process.
HIGH POWER TERAHertz PULSE GENERATION BY RELATIVISTIC LASER PLASMA

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Terahertz radiation has useful features for a number of applications in different fields such as security, communications, medicine and science. Search of high-power terahertz with high conversion efficiency enabled with relativistically intense lasers is an important problem of current interest. High-power terahertz radiation having broadband spectrum was observed in a number of experiments where plasma was produced by short relativistically intense laser pulses. There is known several mechanisms of generation of the THz pulses, e.g. radiation generation during sheath acceleration process at the target rear surface or THz emission by electrical current of fast electrons escaping a target. In this work we compare efficiency of both mechanisms of THz radiation in interactions of short laser pulse with metal targets in a form of foil and wire.

The theory of THz radiation generated due to escape of hot electrons and non-quasineutral plasma expansion into a vacuum has been developed. Contributions of both these effects to the volumetric and surface THz pulse emission have been studied and corresponding conversion efficiencies from laser to THz radiation have been estimated. The dependences of conversion coefficients on laser intensity and laser pulse duration are presented. We also demonstrated that quick surface charging by escaping of laser-heated electrons can be an effective mechanism of surface THz pulse generation in planar and wire targets.

This work was supported by the Russian Foundation for Basic Research (Grant N.16-02-00088-a).
Shortening of the laser pulse towards an optical cycle duration and its focusing on a singlewavelength spot size allows to achieve the maximal intensity in the minimal volume, the lambdacube regime. With only mJ energy at kHz repetition rate such laser systems will make it possible to perform a detailed investigation of relativistic effects with highest field gradients while interacting with solid density target [1]. Applied to overdense plasma targets such laser pulses can lead to efficient production of attosecond electromagnetic pulses [2] and electron bunches [3], which are scalable towards shorter durations and higher energies [4].

In this presentation we will review the recent progress in lasers development, experiments and modeling in this research area.

LASER ENGINEERING OF RELATIVISTIC ELECTRON MIRRORS FROM GAS JET TARGETS

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Engineering of attosecond relativistic electron beams from thin gas jet targets with superintense ultra-short laser pulses is considered. For perpendicular incidence of the relativistic laser pulse at the gas jet, a longitudinal component of the Lorentz force causes a shift of the plasma electrons in the direction of propagation of the laser pulse and can accelerate them to relativistic velocities. If the pulse amplitude is large enough, and the thickness of the gas jet is reasonable, then full evacuation of the electrons from plasma near the laser beam axis takes place. These electrons can form a relativistic electron bunch, which diameter is determined by the diameter of the laser pulse and length is considerably smaller than the laser wavelength. Such a structure can be called a relativistic electron mirror. The threshold thickness of the gas jet is determined by the amplitude and duration of the laser pulse and the initial density of electrons in the gas jet. Requirements on the front duration of the laser pulse for the case of the gas targets are significantly decreased in comparison with the nanofilm targets; besides, the use of the gas targets facilitates creation of installations with high pulse repetition rate. All these features are very important for experiment.

Formation of relativistic electron mirrors from gas jet targets was studied with two-dimensional particle-in-cell simulations. Achievable characteristics of the mirrors were found for different transverse distribution of the gas jet density as well as for the laser pulses of different duration. It was shown that, in all considered cases, it is possible to choose the system parameters (pulse amplitude, density and thickness of the gas jet) in such a way that a single relativistic electron mirror can be formed. Thomson backscattering of a counter-streaming probe laser pulse off such a relativistic electron mirror was also simulated and the parameters of generated coherent ultrashort x-ray pulses were studied.
RELATIVISTIC PLASMA PHYSICS WITH TABLE TOP TW FEMTOSECOND LASERS: SCIENCE & APPLICATIONS

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We present results of the experimental and numerical studies of hot electron generation and high efficiency gamma production by the interaction of relativistically intense laser pulses with solids. Special attention was paid to optimization of the target and laser radiation parameters. We used the Ti:Sa laser system (p-pol, 800 nm, 10 Hz, 40 mJ, 45 ± 5 fs and $I_{\text{max}} = 5 \times 10^{18} \text{W/cm}^2$, ASE & prepulses < $10^{-8}$). For some experiments contrast ratios at the nanosecond and picosecond time scales were adjusted to achieve the desired phenomena. In other experiments the Nd:YAG laser (1064 or 532 nm, 30-120 mJ, 6 ns, $I \approx 10^{12} \text{W/cm}^2$) was used to create the controlled long and dense pre-plasma layer. This laser was locked with the Ti:Sa laser system with accuracy better than 1 ns. Different targets were used: metal plates made of Fe, Mo, W, thick polymer films and plates, as well as different structured targets. We also discussed applications of our studies in nuclear physics, microelectronics, and other areas of research.

NEW ANALYTICAL SOLUTIONS FOR RELATIVISTIC SELF-FOCUSING OF A LASER BEAM

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By using the method of renormgroup symmetries, we present a new approximate analytic solution of a Cauchy problem for the nonlinear Schrodinger equation describing steady-state light beams in a medium with relativistic nonlinearity. This theory involves the self-consistent description of nonlinear variation of the plasma refractive index due to the relativistic increase in the mass of electrons, radial relativistic ponderomotive effect, and laser beam diffraction. A classification of self-focusing solutions is given. The existence of both wave-guide and several types of self-focusing solutions is proved for an entering beam with Gaussian radial distribution of intensity. The found solutions differ in terms of the radial positions where the solution singularities appear.
This work was supported by the Russian Science Foundation (Grant No.17-12-01283).

SELF-GENERATED MAGNETIC VORTICES GROWTH AND PROPAGATION IN INTERPENETRATING ELECTRON-ION BEAMS


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Collisionless shocks generated by colliding relativistic plasmas are studied using particle-in-cell (PIC) simulations. The shock is produced due to the Weibel instabilities that generate current and density filaments and small-scale magnetic fields that are amplified from initial fluctuations.

Localized regions of strong magnetic field in the form of magnetic vortices upstream of the shock are observed in the simulation develop during the nonlinear evolution of the electron and ion filaments. The vortices developing from the merger and subsequent pinching of the small-scale filaments are shown to be moving in the direction opposite to that of the shock. The numerical simulation also witnesses formation of nonthermal energy distribution of accelerated electrons due to stochastic heating in turbulent plasma fields. We present simple analytical estimates that are confirmed by the PIC simulations. This work was partly supported by Russian Foundation for Basic Research (Nos. 15-02-03042, 16-52-50019, 16-02-00088, 17-02-00366).
FAST PONDEROMOTIVE MODIFICATION OF THE ELECTRON DENSITY PROFILE IN LASER ABLATED PLASMA

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Quasy-stationary self-consistent electron density profiles of the inhomogeneous plasma stream (moving parallel to the density gradient) interacting with the electric field of an ultrashort (fs) laser pulse are calculated in the framework of the one-dimensional model with nonlocal polarizability. Because the ions have no time for the ponderomotively-induced modification of their profile during the ultrashort pulse, the electron profiles are supposed to be formed against the background of a given (linear) ion density space distribution and settled down as a result of the equilibrium between the averaged ponderomotive force of laser pulse, the electron pressure, and the static space-charge field under the conditions of a strong violation of quasi-neutrality. The average electron density and the longitudinal (parallel to the density gradient) ac electric field are found to be greatly space-modulated in the region downstream of the plasma resonance point. With the first-order spatial dispersion (plasma-flowing-caused nonlocality) taken into account, the step-like deformation of the electron density profile induced in the plasma resonance region by the ultrashort laser pulse acquires a finite space scale and is followed by formation of a quasi-periodical structure (with the same characteristic scale) after the passage of the plasma flow through the critical density surface. The space charge developing in the case of a fixed profile of the ion density does not prevent formation of the small-scale electron structure, but eliminates the possibility of the extensive plateau of the space-average density. The electron profile modification considered affects greatly the fast particles generation and the resonance absorption in inhomogeneous plasma. The model considered has sufficient commonness and can probably provide a qualitative insight into the deformation structure of the plasma resonance region under various actual conditions of plasma creation (laser ablation of metal targets, optical ionization of metal films, expanding atom clusters, ionized jets, etc.).
AUTORESONANT ACCELERATION OF ELECTRONS BY THE POWERFUL LASER RADIATION

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We report the results of numerical solution of electrons motion equations in the field of ultrashort laser pulse, propagating along the steady magnetic field at the conditions of cyclotron autoresonance [1]. It is shown that in the absence of the condition of cyclotron resonance during injection of electrons they momentarily escape the range of interaction. Laser radiation of the circular polarization is defined in the paraxial approximation in the form of the Gaussian beams of the basic and the first modes. Corrections of the first approximation to the components of the radiation field are taken into consideration [2]. Calculations show that corrections of the first order to the transverse components exert no sufficient influence on the autoresonant motion of electrons whereas the longitudinal components of the first approximation play a major role. It is also shown that the specific form of the pulse is inessential. Images of the spatial distribution of the vectors of the radiation field in the transverse plane depending on the longitudinal coordinate (the direction of the radiation propagation) are obtained. It is shown that the character of changes of energy of an electron beam essentially depends on its position of injection in the focal plane. In this case acceleration as well as deceleration is possible in dependence on the position of injection of the electron beam. It is shown that under the optimal conditions of injection the mechanism of the cyclotron autoresonance can provide sufficiently high efficiency of acceleration of ultrarelativistic electrons in the field of powerful laser radiation with sufficiently high average rate at the distance of the order of two Rayleigh lengths. The basic mode is preferable due to more simple description of that mode, higher acceleration rate and wider acceleration zone of injection of an electron beam.


FROM QUANTUM TO CLASSICAL RADIATION REACTION:
A FOCUS ON STOCHASTICITY EFFECTS

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The development of multi-petawatt laser facilities such as the Apollon laser in France or the ELI project throughout Europe has lead to a regain of interest in the problem of modeling the dynamics of radiating ultra-relativistic electrons interacting with a large amplitude (relativistic) light pulse. This problem known as radiation reaction can be treated in the framework of classical electrodynamics by adding to the Lorentz force a so-called radiation reaction force as proposed e.g. by Landau & Lifshitz. When the energy of the photons emitted by the electrons approaches that of the radiating electron, quantum electrodynamics (QED) effects become important and the stochastic nature of photon emission needs to be taken into account. In this work, we focus on the intermediate regime where the energy of the emitted photons remains small compared to that of the radiating electrons, and yet where the stochastic nature of photon emission cannot be fully neglected [1]. Our approach relies on both linear Boltzmann and Fokker-Planck descriptions of the electron dynamics, and shades new light on this regime of interaction. It allows to capture in a simple way the reduction, due to quantum effects, of the power radiated away by ultra-relativistic electrons through the derivation of a modified radiation friction force. It also allows to account for the stochastic nature of photon emission via the derivation of a diffusion term, and it also provides us with a clear picture (ordering parameters, and characteristic modifications of the radiating electron distribution function) of the transition from the classical to quantum regime of radiation reaction. Last but not least, this approach allows us to propose a simple particle pusher for radiating electrons that can be implemented, e.g. in Particle-In-Cell codes, and that extends their capabilities to the weakly quantum regime without requiring the need of developing Monte-Carlo tools. In this talk, I will present both the theoretical approach and particle simulations to validate it.
The laser-plasma methods of electron and ion acceleration as well as production of secondary radiation attract a lot of attention due to numerous potential applications for astrophysical science, inertial fusion, nuclear physics, material science, biology and medicine. In this report we review recent theoretical researches related to the production of energetic ions and electrons and emission of secondary electromagnetic radiation in the interaction of short relativistically intense laser pulses with different targets. We present theoretical models and 3D PIC simulations on optimization of charged particle acceleration from thin foils and low-dense targets by circularly and linearly polarized laser pulses. An advantage of a new effective scheme of synchronized laser triggered ion acceleration by a slow light pulse of relativistic intensity is discussed. Based on 3D simulation we show optimal laser-target design to get most effective electron source suitable for producing X-ray pulses due to electrons conversion to bremsstrahlung radiation in a converter target. The several theoretical methods of THz radiation due to laser irradiation of thin foils or metal wires are also discussed.

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CHARACTERIZATION OF THE HOT ELECTRON ENERGY DISTRIBUTION
IN LASER INTERACTION WITH METAL TARGETS

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Knowing of the energy distribution of hot electrons generated in high-intensity laser interactions with metal targets is of importance for development of K-alpha sources for diagnostic applications [1] and for inferring conversion efficiencies of laser energy into fast ignition relevant hot electrons [2]. The hot electron temperature deduced from measurements is often compared with the main intensity scaling laws [3, 4]. Two groups of hot electrons were revealed by shadowgraphic [5], K-alpha [6], bremsstrahlung and K-alpha [1, 2] measurements. Average energy of the colder component is less than the scaling predictions. Simulations indicate the possibility of using this component for optimum core heating in cone-guided fast ignition [7].

In our experiment, s-polarized subpicosecond laser pulses of 1.06 μm wavelength were focused on silver targets up to intensity of 2x10^19 W/cm². A two-temperature hot electron distribution was revealed by measurements of the bremsstrahlung emission from a massive silver target by the filter and the knife methods. The latter is usually used to obtain a spatially resolved image of the x-ray emission and then to deduce its spatial extent [6]. To deduce information about the electron distribution, we have developed analytical models of bremsstrahlung and K-alpha emissions, applicable in given experimental conditions. Absolute K-alpha measurements were used to determine conversion efficiency of laser energy into the energy of hot electrons.

The temperature of the colder component (T₁ = 140 keV) was defined by fitting the experimental and modelling data at the fixed temperature of the hotter component, determined according to the ponderomotive scaling [3] (T₂ = 1.66 MeV). Obtained temperatures correspond to the results of 2D PIC simulation of hot electron generation in cone-guided approach to fast ignition [7]. Conversion efficiency of laser energy into the energy of hot electrons with the two-temperature distribution is less than 2%. Approximately 90% of absorbed laser energy is contained in the colder component. These results are confirmed by comparison of the K-alpha yield, modeled using obtained hot electron spectrum, and measured K-alpha yields from a silver foil, deposited on bulk substrates. Estimated mean number density of the colder electron component is about the subcritical density. The density of the hotter component is about two orders lower. A possibility to analyze parameters of the hotter electron component taking into account errors of the measurements is under discussion. Assumption about one-temperature parametrization of the hot electron spectrum with the slope temperature described by the ponderomotive scaling [8-10]
strongly overestimates conversion efficiency of laser energy into the energy of the hotter electron component.

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References

SOFT X-RAY RELATIVISTIC HARMONICS AND SOFT AND HARD X-RAY FEL BEAMS HIGH-PERFORMANCE IN SITU IMAGING BY LIF DETECTORS WITH SUB-MICRON RESOLUTION


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Review of results, obtained by using recently proposed new approach for applying a color center formation in LiF crystals and films to perform a high quality, single-shot in situ imaging of the near and the far field intensity distribution of soft X-ray lasers (SXRL), soft X-ray free electron lasers (SFEL), soft X-ray relativistic high order harmonics (HOH) and hard X-ray free electron laser (XFEL) beams will be presented. Provided experiments demonstrated that such technique allowed to obtained images in a single-shot with of ~ 700 nm spatial resolution for soft X-ray spectral range and ~ 1.0 µm for hard X-ray photons with energy of 10.1 keV. We demonstrated that sub-micron resolution, high dynamic range LiF detectors allowed applying a single shot imaging technique for characterizing relativistic high order harmonic (HOH) beams, generated by an oscillating electron spikes formed in underdense gas jet plasma irradiated by relativistically self-focusing laser pulses and measure the size of single emitting plasma zone, which have been estimated as smaller as of 200 nm. Additionally the LiF film was used for near-field diffraction imaging of the HOH beam and investigation of its coherent properties. The focusability of the HOH beam was investigated using spherical Mo/Si mirror and LiF crystal. Using such technique the sizes in the scale of 200 – 900 nm and complexity of the HOH source structure have been measured. It was shown experimentally and by modeling that diffraction of SXRL beams on the periodical structures and registration of diffracted patterns by LiF X-ray detectors opens new possibility of measuring the intensity, coherence and spectral distribution across the full profile and in local areas of SXRL beam with accuracy of 10 - 20 %. High sensitivity and large dynamic range of the LiF crystal detector allowed measurements of the intensity distribution of the hard X-ray SACLA XFEL beam at distances far from the best focus as well as near the best focus. In situ 3D visualization of the SACLA XFEL focused beam profile along the propagation direction is realized, including propagation inside photoluminescence solid matter.
BETATRON RADIATION OF CHARGED PARTICLES IN LASER-PLASMA ACCELERATORS

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In a laser-plasma accelerator, relativistic particles move under the action of wake fields generated in a plasma by a short laser pulse and undergo betatron oscillations. As a result, the synchrotron radiation is emitted. This radiation can be used for many applications [1], but can affect also characteristics of the electron beam [2]. A model for numerical simulation of the acceleration of polarized electrons has been elaborated in Ref. [3].

In this work, to describe the synchrotron radiation of particles the radiative reaction force in the Landau-Lifshitz form was used and quantum recoil effects were taken into account [4]. The spin precession dynamics of electrons was determined by the Thomas–Bargman–Michel–Telegdi equations [5]. In the prescribed conditions, the critical energy of the emitted photons was estimated and their influence on the electron trajectory and beam polarization dynamics was studied.

In the simulation the wake field was generated by a short high-intensity laser pulse with a duration of 0.5 ps, wavelength of 1 μm and total energy of 100 J in a preformed plasma channel with a parabolic initial density profile of plasma electrons. Considered subpicosecond intense laser pulse corresponds to the laser system PHELIX (Petawatt High Energy Laser for heavy Ion eXperiments) at GSI, Darmstadt [6].

HOT ELECTRON GENERATION IN INTERACTION OF INTENSE LASER PULSES WITH NEAR-CRITICAL PLASMA

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Studies of the laser-plasma interaction with targets of various types were conducted by 3D PIC simulations for future experiments optimization. Characteristics of hot electrons ejections were analyzed in detail. Laser pulse parameters in simulations corresponded to the parameters of the PHELIX laser facility at GSI, Darmstadt. The scheme of the normal incidence of the laser pulse onto the target was considered in the work.

Firstly, the simulations were performed with an overcritical plasma layer having the uniform density distribution. These calculations did not consider the influence of the pre-pulse. The results of the simulations are in a good agreement with existing theoretical concepts on the interaction of relativistic femtosecond intense laser radiation with the sharp boundary of the overcritical plasma density. Energy spectra of accelerated electrons are close to the ponderomotive scaling for the incident laser amplitude.

Secondly, overcritical targets having pre-plasma on its surface with various scale lengths were considered in the work. Pre-plasma density profiles have been previously obtained by a two-temperature hydrodynamic modeling [1]. The results demonstrate the increase of the number of hot electrons in comparison with the case of the uniform overcritical plasma layer due to absorption mechanisms presence in the near-critical part of the plasma density profile.

Finally, the study of interaction with near-critical plasma layers having the uniform density distribution was conducted. These targets can be obtained in experiments when the laser pre-pulse is applied to the foam. Electron densities of foams in the range 0.5–1.0 of the critical density and thicknesses in the range 100–500 µm were considered. It is shown that heating of electrons mainly occurs under the action of the ponderomotive force of the laser pulse. The amplitude increases up to three times during the interaction because of the self-focusing effect in underdense plasma [2]. Obtained energies of accelerated electrons can be approximated by the Maxwell’s distribution with the temperature 8.5 MeV. The charge carried by electrons with energies higher than 30 MeV is about 30 nC, that is 3–4 order of magnitude higher than the charge predicted by the ponderomotive scaling for the incident laser amplitude.


INITIATION OF SELF-SUSTAINED QED CASCADES IN GENERIC ELECTROMAGNETIC FIELD OF ELECTRIC TYPE

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Generation of self-sustained (or A-type, [1]) QED cascades is one of intriguing QED phenomena that are anticipated to become observable under laboratory conditions during the next decade due to awaited forthcoming ultra-high intensity laser facilities. Possibility of initiation of such cascades strongly depends on both field strength and structure, and is quantified by the value of so called quantum dynamical parameter $\chi$ of particles involved in laser driven dynamics (see e.g. [2,3] for details). This parameter can be interpreted as a local field strength in units of Sauter-Schwinger field $E_S$ in the proper reference frame of an electron. Thus if initially slow seeding particles with $\chi \ll 1$ start accelerating in laser field so violently that $\chi$ grows up to $\chi 1$ during time interval shorter than laser period then A-type cascade can occur.

We present a general expression for time dependence of the key parameter $\chi$ for a seed electron in a slowly varying strong ($a_0 \gg 1$) electromagnetic field of electric ($E^2 - H^2 > 0$) type on a time scale $1/a_0 \omega \ll t \ll 1/\omega$. We show that at this time scale $\chi$ is always growing as time squared while the coefficient is determined by local field configuration. Our result generalizes a few known expressions of this sort for various particular field models, e.g. the rotating electric field [5], circularly or linearly polarized standing waves [4] and multiple focused beams [5]. The obtained general expression for $\chi(t)$ allows to generalize the condition of QED cascade initiation that was originally conjectured in [2,3].

In Ref. [5] it was proposed that in order to maximize cascade multiplicity for a given laser power one should search for field configurations that maximize growth rate of $\chi$. In context of that idea our general expression can be useful in further searches for optimized field configurations favorable to design future experiments by reducing such critical requirements as e.g. the net laser power.

ON EMITTANCE OF ELECTRON BUNCHES ACCELERATED IN GUIDING STRUCTURES

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Short (femtosecond) intense laser pulses propagating in the guiding structures (plasma channels and capillary waveguides) make it possible to achieve high accelerating wakefields (of the order of $10^8$ V/cm) in the plasma. These fields can be effectively used for creation of compact accelerators of electrons with accelerating gradients that are much higher than in the traditional radiofrequency accelerators.

The possibility of practical use of electron bunches accelerated in wakefields behind laser pulses propagating in a plasma is determined by the characteristics of the bunches, such as their average energy and energy spread, charge and emittance. Ensuring a low emittance value is crucial for the concept of a multistage electrons accelerator.

The growth of emittance of electron bunches accelerated in the guiding structures was investigated [1]. It is shown that the emittance is determined substantially by longitudinal and transverse sizes of the accelerated electron bunch, its phase of injection into accelerating wakefields and by the accuracy of laser pulse focusing into a guiding structure, which determines the quality of accelerating wake fields. In particular, it is shown that the low emittance requirement imposes an order of magnitude more severe restrictions on the accuracy of laser pulses focusing into guiding structures than the requirements of high energy gain, of low energy spread and the conservation of a large number of trapped and accelerated particles in an accelerated electron bunch.

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ION ACCELERATION IN ADIABATION EXPANSION OF CYLINDRICAL MULTISPECIES PLASMA

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Recently, considerable interest has been focused on study of radial ion acceleration in relativistically strong ultrashort laser pulse and plasma interaction. Non-Maxwellian ion energy spectra were observed experimentally including monochromatic ion group formation. Here, we pay attention to the theory of adiabatic multispecies radial plasma expansion triggered by laser-heated electrons. This mechanism is different from ponderomotive ion acceleration that works for longer laser pulses with duration exceeding inverse ion plasma frequency. The analytic solution of the Cauchy problem is constructed for a system of kinetic equations for the hot cylindrical electron-ion plasma in quasi-neutral approximation. It can be applied to description of the acceleration of ions caused by the hot electrons generated by a laser beam which propagates in the transparent medium and produces laser-plasma channel. The solution is found under conditions where the Debye radius of the electrons is considerably smaller than the characteristic laser beam radius. Our theory is based on the innovative renormalization-group method. The temporal and spatial dependences of the distribution functions of particles were obtained and their integral characteristics, such as density, mean velocity, temperature, and energy spectrum were found.

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OPTIMIZATION OF LASER PULSE- LOW DENSITY PLASMA INTERACTION

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At the present time there are some techniques of efficient electron acceleration by short laser pulses, including laser-wakefield acceleration. They allow to reach the high collimation of high-energy electron beams, but the total charge of such beams turns to be rather small. PIC-sumulations of laser pulse interaction with low-density plasma have been carried out to optimize plasma characteristics for achieving the maximum charge of electrons with energy exceeding 30 MeV and the maximum energy of corresponding X-rays. In our calculations the energy of the laser pulse has been chosen as a few Joules with the peak intensity being $10^{21}$ W/cm$^2$. 
EFFECT OF THE LASER PULSE SELF-FOCUSING ON THE INJECTION AND ACCELERATION OF ELECTRONS IN THE PLASMA WAKE FIELDS

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The wake plasma waves generated by high intensity laser pulses are of interest for acceleration of electrons to high energies. In particular, the creation of high-energy electron sources for injection into a laser-plasma accelerator was studied in [1], [2]. The aim of this work is to study the effect of self-focusing of a laser pulse on the injection and acceleration of electrons in the plasma wake fields. The laser pulse energy (40 mJ) and the plasma density distribution were selected in accordance with the experiment [2]. Calculations were performed using the three-dimensional "particle-in-cell" method [3] with the following parameters: the length of the laser pulse at half maximum equals to 15 µm (45 fs), the focal spot radius at the level 1/e² equals to 8.4 µm, the maximum intensity at the focus equals to $9.4 \times 10^{17}$ W/cm². The density distribution of the hydrogen plasma is Gaussian, with the density maximum $n_{\text{max}} = 6.6 \times 10^{19}$ cm⁻³.

At laser pulse power exceeding the critical value by a factor of three, self-focusing of the laser pulse and steepening of the pulse front are observed. These processes lead to self-modulation instability, generation of a wake plasma wave, to injection and acceleration of trapped electrons. The maximum energy of accelerated electrons approaches 8 MeV that agrees with data obtained in the experiment.

NONLINEAR PLASMA RESONANCE IN INHOMOGENEOUS RELATIVISTIC PLASMA

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Based on our theory of relativistic plasma resonance via the renormgroup symmetries method, we have presented a theoretical investigation of the harmonic generation, quasistatic fields, and resonance absorption of laser radiation in an inhomogeneous laser plasma. The spatial-temporal and spectral characteristics of the stationary radiation field is described. The spectral analysis points to the existence of a power law spectrum for the radiation field. The total reflection coefficient of the laser radiation is calculated, as well as at the fundamental frequency. The obtained results show that the relativistic effects should be taken into consideration already at moderate intensities of $10^{12}$-$10^{15}$ Wt/cm, which suggests that the results obtained using the weakly nonlinear and nonlinear nonrelativistic theories of the plasma resonance should be partially reviewed and corrected.

INJECTION AND CAPTURE OF ELECTRONS IN WAKEFIELDS GENERATED BY A RELATIVISTIC ION BUNCH

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Experiments on ion acceleration driven by high intensity laser pulses over the past ~15 years have demonstrated the generation of multi-10s of MeV proton and ion beams with remarkable properties. The unique exploratory mission of this research is to build the scientific foundation needed to develop high energy laser-particle accelerators, to expand the fundamental understanding of matter at very high temperature and density conditions. This presentation will discuss the ion acceleration obtained on 1.5 PW laser. The measurements provided for the first time the opportunity to extend scaling laws for the acceleration process in the ultra-short regime beyond the $10^{21}$ W/cm$^2$, and to access new ion acceleration regimes. The newly found scenarios of ions acceleration offers more favorable proton energy scaling with laser intensity than known so-called Target Normal Sheath Acceleration mechanisms [1]. We have found that in this regime of ion acceleration particles spectrum can be steering by controlling the contaminant layer. “Monoenergetic” ion beams is obtained with appropriate ratio of heavy and light ion species in the contaminant layer. However, in the PW laser-solid interaction experiments the dynamics and the properties of the target surface can result in significant amount of reflected back laser light which can be “re-collimated” by the focusing parabolic mirror and propagate back into the laser chain. At high laser pulse contrast on the laser-target interface a regular structure is generated during the interaction which acts as a grating and some of the diffraction maximum will be under back reflection angle. This back reflected light of the order of 1% of the incident laser energy can have serious consequences not only on laser (cause serious damage) but also it can affect the interaction conditions [2]. 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. Our findings pave a way to achieving an ion source and beam desire parameters and they encourage further development of laser plasma-based accelerators.

2. Ultrafast phenomena in condensed matter and ionized gases

Chair S.A. Uryupin

LASER INDUCED ULTRAFAST SWITCHING PROCESSES IN DIELECTRICS FOR APPLICATIONS IN NANOPHOTONICS

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The electron dynamics in wide band gap materials such as diamond subjected to intense ultrashort laser pulses is studied theoretically for a wide range of pulse energies (1-103mJ/cm\textsuperscript{2}) and laser wavelengths 515, 800, 1030 and 1550 nm. The feasibility of diamond for sub-femtosecond (attosecond) pulse generation with emphasis on the mechanisms of high harmonic generation (HHG) in solids \cite{1, 2} will be discussed. Aspects of the linear response of photo excited plasma and the change of optical properties in terms of transient dielectric function that represents the gradual transformation of diamond bulk into a plasmonically-active phase supporting photoexcitation and propagation of surface plasmon-polaritons (SPPs) \cite{3} will be presented. Thus an integrated fundamental qualitative and quantitative picture of the interaction of ultrashort pulses with high refractive index materials is obtained providing calculations of their dielectric constant prompt modulation in the ultraviolet (UV)-visible (VIS) to near-infrared (NIR) spectral range. This allows to predict and potentially extend the capabilities of the dielectric nano-optical meta devices and the photo-modulated meta surfaces based on them. These studies may uncover ultimate limits of the high-index dielectric materials for their potential applications in high harmonic generation, ultrafast optical switching, spatial phase modulation and saturable absorbers.

\begin{itemize}
\item \textsuperscript{1}S. Lagomarsino et al., Phys. Rev. B 93, 085128 (2016).
\item \textsuperscript{3}T. Apostolova et al., arXiv preprint arXiv:1701.04650 (2017).
\end{itemize}
ULTRAFAST ELECTRON MICROSCOPY AND DIFFRACTION WITH SUB-100-KEV ELECTRON PULSES

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The study of the structural dynamics of various materials with a high spatial and temporal resolution is one of the key lines of research. Ultrafast electron microscopy and electron diffraction are rapidly developing fields of modern science. These methods are based on probing coherent laser-induced processes in various materials using photoelectron bunches. This approach allows one to supplement the high temporal resolution provided by modern pulsed lasers with the atomic spatial resolution, inherent in diffraction methods. As a result, one can make a video of the behaviour of a material under study, matched in the four-dimensional space – time continuum. In our talk we will discuss our experimental activity in this field. This study was supported by the Ministry of Education and Science of the Russian Federation (Project No. RFMEFI61316X0054).

DENSITY-FUNCTIONAL SIMULATIONS OF STRONG-FIELD PHYSICS

IN SOLID SLABS

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Strong-field physics effects in laser-atom interaction, such as energetic electrons in photoelectron spectra, high-harmonic generation or nonsequential multiple ionization can be largely understood in terms of classical electron orbits revisiting the ion. This paradigm has been recently applied to nondestructive laser-solid interaction successfully as well, opening up a new era of strong-field physics with (relatively) weak lasers and exciting potential applications in the field of ultra-fast, light-driven electronics and excitonics.
The qualitatively new aspect in solids clearly is the band structure. Electronic motion in the conduction band replaces the electron motion in the continuum with simple dispersion relation \( p \sqrt{1/2m} \) in the atomic case. Further, the usually adopted single-active electron (or hole) approach appears to be less adequate in the solid case. Nevertheless good agreement between a classical trajectory viewpoint and experiment and (single-active electron) solutions of the time-dependent Schrödinger equation have been reported [1].

In our presentation, we introduce a one-dimensional model of a many-electron solid slab interacting with a laser field and solve for the time-dependent, non-perturbative dynamics using time-dependent density functional theory in local spin density approximation. We are then able to compare single-active electron results for harmonic generation and other processes with the corresponding all-electron results. Further, we can freeze and unfreeze the effective Kohn-Sham potential in order to determine the importance of electron-electron interaction. Finally, the simulation of a finite solid slab instead of bulk allows to identify surface effects. We find that the single-active electron response of the uppermost valence electron alone is rather different from the full many-electron response, rendering single-active electron models doubtful. Still, simple trajectory models can qualitatively explain the calculated time-frequency (Garbor) spectra. A frozen Kohn-Sham potential turns out to be a good approximation in the laser intensity regime considered.


**ON THE MECHANISMS OF THZ RADIATION GENERATION ON METAL SURFACES**

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We present time-domain solution of the equations describing low-frequency radiation generation when metal surface is irradiated by a non-damaging femtosecond optical pulse of s-polarized radiation. Ponderomotive force, drag current and temperature gradient along the surface are found to be sources of the THz radiation. Comparison of these sources under typical experimental conditions reveals that both drag current and thermal gradient give similar contribution to the THz signal. Ponderomotive force effect is several times less than these two, and exists only during the incident pulse.
INTERFERENCE STABILIZATION OF ATOMS IN STRONG LASER FIELD AS A WAY TO OBTAIN AMPLIFICATION AND LASING IN DIFFERENT FREQUENCY RANGES

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It is known from [1, 2] that the influence of high intensity laser fields causes the significant reconstruction of atomic system resulting in stabilization phenomenon, i.e. population trapping in high-lying Rydberg states. One of the possible ways to study the structure of such reconstructed (dressed) by the laser field quantum systems is to analyze their spontaneous emission. We have developed the new approach how to introduce spontaneous emission into the Schrödinger equation [3]. The proposed approach is applied to examine the dynamics of field-driven atomic system in the regime of interference stabilization (IS). Main predictions of IS theory are testified and confirmed. The effect of IS is also proposed to create the plasma channel with population inversion between set of Rydberg states and the ground state for the conversion of the input laser energy into the VUV and XUV frequency band [4]. Furthermore, there is a possibility to create a population inversion between high-lying Rydberg states which can be used for lasing and amplification in the IR, mid-IR and sub-terahertz frequency band.


Ultrashort-pulse lasers are an excellent tool for processing of any kind of materials. By proper choosing the irradiation conditions, it is possible to create highly-localized surface modifications with minimal heat-affected zone due to minimized heat diffusion effects. The interaction of ultrashort laser pulses with surfaces involves a wealth of the physical processes, depending on the material kind and laser light properties. Even within the same material family, considerably different modifications can be achieved when applying similar laser pulses. Improvement in understanding of the physical mechanisms of laser-induced material modification/ablation and contributions of individual processes is of vital need for further advance of ultrafast laser material processing techniques for a broad range of applications, including integrated optics and photonics, optofluidics, optomechanics, optoelectronics, surface micro- and nanostructuring.

The optical response of metal surfaces to high-power laser excitation (HPLE) is one of important topics of laser-matter interaction which remains to be poorly understood. It is known that certain metals, which are highly reflective at normal conditions, become considerably absorbing during the HPLE stage. The amount of the absorbed energy is a key parameter which determines the post-irradiation evolution of the material. Numerous optical models have been proposed to describe this effect and to link the actually absorbed laser energy with the incident laser energy for direct comparison of the simulated material dynamics and the experimental irradiation conditions. This includes variations of the Drude and Drude-Lorenz models, contribution of plasma-like response of free electrons in metal, as well as attempts of ab initio simulations of the optical response.
In this talk, the existing models of the optical response of metals will be overviewed with critical assessment of their applicability to the conditions of HPLE, based on direct comparison with experimental data for gold and zinc. The results of two-temperature modeling (TTM) of femtosecond laser irradiation of the two metals by the temporally and spatially Gaussian laser pulses with integration of the reflected beam part over the irradiation spot will be reported in comparison with the experimentally measured reflectivity as a function of laser fluence. Different models of reflectivity combined with the TTM modeling will be compared. The effects of the physical properties of laser-excited metals on their optical response under HPLE conditions will be analyzed. It will be demonstrated that an optical model itself cannot describe the experimentally measured data on reflectivity without accurate knowledge about dynamic behavior of thermophysical properties of metal upon laser excitation. Some implications on mutual impact of dynamically

SURFACE PLASMON POLARITONS: THEIR PROPERTIES FOR DIFFERENT METALS AND THE CONSTITUTIVE ROLE IN ULTRAFAST LASER PROCESSING

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Irradiation of metals and semiconductors by intense ultrashort laser pulses leads to a transient change of material optical properties, subsequently inducing a strong increase in optical absorption (losses) [1] and creating favorable conditions for excitation of several types of surface electromagnetic waves (SEW) [2,3], including Surface Plasmon Polaritons (SPP) [3-5]. The classical SPP theory was developed for noble metals under the assumption of small perturbations of the free-electron gas and weak plasmonic damping [6]. For intense laser irradiation of materials resulting in out-of-equilibrium conditions, the theory is to be revised by taking into account high optical losses resulting in strong plasmonic damping. Contrary to traditional plasmonic applications, upon intense laser irradiation of material surfaces even small coupling efficiencies are sufficient to provide a spatial modulation of the deposited laser energy, yielding in the formation of laserinduced periodic surface structures (LIPSS) [2-5]. In Ref. [7], it was predicted
theoretically that SPP can efficiently be generated on the surfaces of a wide range of irradiated metals, thus broadening the range of plasmonic applications. The SPP properties such as their spatial period and the decay length were calculated for metals where the optical losses are considerable. In this work, simulations have enabled to classify plasmonically active metals into several groups in respect of the SPP decay length $LSPP$. To check the effects of highly non-equilibrium heating of free electrons at ultrashort laser irradiation, the temporal variations of the dielectric permittivity were modeled numerically for titanium and molybdenum as the examples. For these aims, the two-temperature model was supplemented by the computation of the optical properties in the frame of the Drude model. It has been shown that the SPP decay length is decreasing upon laserinduced excitation of these two metals. Physical insights into SPP propagation indicate that preferably materials with short SPP decay lengths, of the order of 10 $\mu$m, allow the imprinting of highly-regular periodic structures on material surfaces via coherent linking effects in laser scan processing. This theoretical result was successfully verified experimentally for several metals when the laser processing parameters are suitably chosen [8].

REFERENCES

GENERATION OF SURFACE WAVES BY A DRAG CURRENT

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The excitation and propagation of surface waves in the conductors are studied for a long time. A significant number of experimental and theoretical papers are devoted to the study of the features of wave excitation on the conductor surface containing various types of nonuniformities. In the case of smooth conductor surface the excitation of surface waves is possible if nonlinear polarization of the conductor in the localized field of laser pulse is taken into account. The present Communication is devoted to the consideration of the new nonlinear optical phenomenon—generation of surface waves by a drag current appearing at an inclined incidence of a femtosecond laser pulse which is focused by a cylindrical lens. Since the effective frequencies of electron collisions for typical metals at room and higher temperatures are relatively high, the above mentioned mechanism of terahertz surface wave excitation is more efficient than the ponderomotive mechanism. The spectral and energy parameters of the excited surface waves are studied. The total energy of surface waves is calculated and its dependencies on the pulse duration and the focal spot size, as well as the parameters of the conductor, are studied. It is shown that the energy of surface waves is maximal in the case of laser radiation incident almost along the surface of the conductor. The generated waves have terahertz frequencies and their total energy increases with an increase in the effective frequency of electron collisions.
CONVERSION OF TRANSPARENT DIELECTRIC BY INTENSE SHORT LASER INTO TRANSIENT OPTICALLY INHOMOGENEOUS STATE (DIE-MET)

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Abstract

The intense short laser pulse transforms the initially transparent dielectric into electrically inhomogeneous state early in the pulse time [1]. The permittivity of excited material starts to vary in time and in space, and its real part changes from positive to negative values while the mass density remains homogeneous. As a result the unconventional transient material state is created, which combines the dielectric and metal properties simultaneously (we coined it as “Die-Met”). The interplay between the gradient of transient permittivity and the polarisation of the incident laser becomes the major process of the new interaction mode. In a particular relation between the polarization and the permittivity gradient the incident field amplitude grows up while the wave is approaching to the surface where the real part of permittivity turns to zero. That results in the local increase in the absorbed energy density. The complex 3D structure of the permittivity makes a transparent part of excited dielectric (at \( \varepsilon_0 < \varepsilon_{re} < 0 \)) optically active. The electromagnetic wave passing through such a medium gets twisted trajectory and accrues a geometric phase [2]. The plane of polarisation rotation and phase depends on the 3D permittivity structure [3]. Measuring the polarisation and phase of the probe beam allows quantitatively identify this new transient state, which exists around one nanosecond after the end of the hundred-femtosecond-long pump pulse. Note that the unusual transient state of the ultrashort pulse excited Bi was experimentally observed to exist up to 4 ns after the excitation with a 60 fs pump pulse [4], but no explanation for its properties has been given so far. The transient Bi phase exhibit optical properties different neither from those for solid and liquid state, nor from the mixture of liquid-solid states. We discuss the revelations of this effect in different experimental situations and its possible applications.

References


**VOLUME NANOGRATING FORMATION IN LASER-SILICA INTERACTION AS A RESULT OF THE 1D PLASMA-RESONANCE IONIZATION INSTABILITY**

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The formation of periodic nanostructures (“nanogratings”) inside the volume of the transparent dielectric by the series of the femtosecond laser pulses of the moderate intensity has attracted considerable attention in the last two decades as a perspective method for the high density optical information writing and storage. Nevertheless, the physical mechanisms responsible for the creation of the observed self-organized bulk nanostructures are still under discussion. In the report presented we show that the main mechanisms governing the periodicity of the ionization structure in every unit laser pulse (that determines ultimately the periodicity in the induced nanograting) can likely be understood and described in a consistent and noncontradictory manner within the framework of known approaches developed in the studies of small-scale (subwave) ionization-field instabilities of electromagnetic waves in gases. Based on linearization procedure of the equations describing the evolution of electric field amplitude and plasma density, we have analyzed the initial stage of the small-scale ionization-induced instability developing inside the fused silica volume exposed to the femtosecond laser pulse and have calculated the spatial spectra of the instability with the electronhole diffusion taken into account for the first time. This stage is found to results in the formation of some hybrid (“diffusion-wave”) 1D structure with the spatial period that is determined as the geometrical mean of the laser wavelength and characteristic diffusion length of the process considered. Near the threshold of the instability, this period occurs to be approximately equal to the laser halfwavelength in the silica, close to the one experimentally observed.
LASER ABLATION: REVIEW OF SOME RESULTS

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The report reviews our recent results concerning laser-matter interaction in the range of moderate intensities. Cohesive properties of condensed matter are still significant in this range. The report is based on papers [listed below]. Together with used physical models, the set of unsolved problem connected with these results will be presented and discussed. Our work is supported by the grant of the Russian Science Foundation 14-19-01599.

SURFACE NANOSTRUCTURING OF SEMICONDUCTORS IN THE REGIME OF HIGH-DENSITY ELECTRONIC EXCITATION

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In this talk, surface nanostructuring of semiconductors in the regime of high density electronic excitation (above Mott transition and below ablation threshold) will be considered. The examples of ZnO and TiO2 monocrystals and thin films will be presented. Grooves, ripples, spikes, random volcano-like craters, quasi-regular interconnected nanoholes were observed depending on the irradiation conditions. The nanostructuring sensitively depends on the material mass density, crystalline planes orientation with respect to laser polarization, laser fluence and dose. The mechanisms of the structuring will be discussed.
ULTRASHORT HIGH-POWER OPTICAL PULSES FROM SEMICONDUCTOR LASERS BY ENGINEERING PULSED ELECTRICAL PUMPING

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Compact and stable sources of ultrashort high-power optical pulses in ~1 μm wavelength range are desirable for many applications including range-finders for autonomous vehicles, free-space optical communication, two-photon microscopy, nonlinear frequency conversion, arbitrary form signal generation, etc. [1-3]. Such pulses are typically generated by Q-switched or mode-locked solid-state lasers, which are bulky and less efficient comparing to semiconductor lasers. Direct application of laser diodes is hindered by typically low power achievable with these light sources in Q-switched or mode-locked operation. Therefore an interesting alternative is utilization of gain-switched laser diodes. At the same time, high-power short-pulsed pumping of semiconductor lasers required for generation of picosecond-scale optical pulses rise very complex dynamical effects including non-temperature gain clamping [4], power dropout [5], turn-on delay and switching of lasing between the quantum states [6,7] etc.

In this work we experimentally study electrical-to-optical peaking of single- and multi-mode edge-emitting laser diode pulses. The lasers were pumped by high-amplitude electrical pulses of duration below 5 ns and rise-time < 1 ns. It should be noted that lasers of both types were standard commercial single-section devices with no means for short-pulse or high-frequency operation and without saturable absorber. It was found that the thoughtful engineering of the pumping pulse parameters can lead to very significant electrical-to-optical pulse peaking with all studied semiconductor laser samples of both single-mode and multi-mode geometries. With optimal pumping pulse shape, the peaking factor reaches 10-100 values without considerable decrease of the peak output power. For single-mode semiconductor lasers under ~1 ns electrical pumping the optical output pulse peaking down to 35 ps has been demonstrated.

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References:
INFLUENCE OF DEEXCITATION PROCESSES ON THE DYNAMICS OF LASER-EXCITED ARGON CLUSTERS

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The excitation of atomic clusters by intense infrared laser pulses leads to the creation of highly charged ions and to the emission of energetic photons. These phenomena, which follow from ionization processes occurring in the cluster, depend significantly on the population of ground states and excited states in the laser-produced nanoplasma. This makes it necessary to account for collisional excitation and deexcitation processes. We investigate the interaction of femtosecond laser pulses with argon clusters by means of a nanoplasma model. Considering laser-excitation with single- and double-pulses, we analyze the role of excitation and deexcitation processes in detail, and calculate the yield of highly charged ions and of energetic photons in different wavelength regimes. For clusters with initial radii $12 \leq R_0 \leq 35$ nm, the temporal evolution of different plasma parameters was simulated including the number density of the free electrons, the electron temperature, and the population of different ionic charge states. Special emphasis was placed on the effects stemming from the consideration of excitation and deexcitation processes, which are important in order to correctly describe the population of excited states and the associated photon emission. By taking these processes into account in an extended nanoplasma model, we calculated the photon yield from laser-excited clusters. After introducing the different physical processes of the model and their theoretical description – such as tunnel and collisional ionization, the lowering of the ionization energies, the heating, and the expansion of the cluster – numerical results for the ionization dynamics were discussed in the case of the ground state model, in which excited states were neglected. In a second step, excited states were incorporated in the model. To this purpose, individual energy levels were bundled into combined states within a simplified level scheme. Excitation cross sections were calculated and collisional excitation rates were found via an averaging with respect to the electron distribution function and over one laser cycle, whereas the rates for collisional deexcitation were obtained by means of a relation between excitation and
deexcitation cross sections. It was found that with the inclusion of excited states, the ionization dynamics is accelerated, and higher ionic charge states are reached at the end of the laser-cluster interaction. The incorporation of excited states leads to a reduction of the electron temperature because the free electrons lose energy in excitation processes. Moreover, the free electron density is increased due to the enhanced ionization dynamics. For smaller clusters, a second Mie resonance is observed, resulting in a larger heating and a faster expansion. The population of ionic charge states is only slightly modified by deexcitation processes. However, these processes play an important role for the population of ground states and excited states, which in turn significantly determines the emission of radiation from laser-excited clusters. Within a double-pulse excitation scheme, a maximization of a certain ion yield was found to appear at a certain delay time in the case of a smaller pre-pulse followed by a larger main pulse. Here the inclusion of excited states resulted in a flattening of the curves, i.e., the region of “optimal“ delay times was broadened. By assuming a Boltzmann distribution for the individual levels within the combined states, the emission of radiation was investigated. The photon yield from the excitation of outer-shell electrons was computed, and by introducing additional particle species with a K-shell vacancy, also the emission from inner-shell excitations was considered, showing a qualitatively good agreement with other theoretical as well as experimental results.

LASER-INDUCED PERIODIC SURFACE STRUCTURE FORMATION:
NUMERICAL INVESTIGATION OF THE TEMPERATURE DYNAMICS IN METALS AND DIELECTRICS AND SUBSEQUENT STRESS BUILDUP

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In this work, to get insight into laser-induced periodic surface structure (LIPSS) formation, the relaxation of a modulation in the temperature profile is investigated numerically on surfaces of two different kinds of materials (metals and dielectrics) upon irradiation by ultrashort laser pulses. The temperature modulation is assumed to originate from the interference between the incoming laser pulse and the surface-scattered electromagnetic wave, which is considered as the main mechanism lying at the basis of LIPSS formation [1]. The interference causes the periodic
deposition of laser energy into the surface layer of materials, which, in its turn, can trigger periodicities in stress distribution, melting, and ablation, before the final heat dissipation [2,3]. Hence, it is important to investigate the whole route of evolution of laser-irradiated matter from its excitation at ultrashort time scale to the final structure imprinted on material surface.

For comparative studies of the dissipation dynamics of laser energy periodically deposited on material surfaces (fused silica and gold as examples), a 2D model [2] is used. It is based on the twotemperature model (TTM) and considers mechanisms of laser light absorption and energy dissipation specific for the two materials under study. The TTM is coupled with the Drude model of optical properties, considering the evolution of the free-carrier density and/or temperature [4]. The stress build-up is estimated from the local temperature gradients evolution.

The development and decay of the lattice temperature modulation, which can govern the LIPSS formation, is followed during electron-lattice thermalization time and beyond. It is shown that strong temperature gradients can form along the surfaces of both metals and dielectrics within the fluence range of LIPSS fabrication in experiments. The differences in the resulting stresses at the surfaces of gold and fused silica are presented, which can contribute to the periodic pattern formation. Considerable changes in optical properties are found as a function of time for gold, for which a constant reflectivity is usually assumed. The effects of the nonlinear absorption on the surface temperature dynamics and stress formation are will be discussed.

FRAGMENTATION OF LIQUID METAL MICRO-DROPLETS BY LASER-GENERATED SHOCKWAVES


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Laser-generated shockwaves are widely used in experimental studies of the rupture and fragmentation of condensed matter. Studies of laser-generated shocks in solid materials have been conducted for a rich variety of materials and in broad spectrum of experimental conditions. For liquids, most of the studies on laser-generated shockwaves have been performed only for optically transparent substances, e.g. water. The negligible attenuation of visible light in such materials enables direct visualization of the liquid matter rupture. In contrast, studies for liquid metals are scarce. Experiments with liquid metals are, technically, more sophisticated, due to the requirement to operate at elevated temperatures. However, an understanding of the response of liquid metals, subjected to high-power laser pulses, is crucial for applications, such as laser ablation through spallation or target optimization for laser-produced plasma sources that emit extreme ultraviolet radiation for lithography.

We report on experimental studies of the deformation and fragmentation of free-falling liquid metal droplets by Mbar-scale shockwaves that were generated by high-intensity ultrashort pulses of Ti:sapphire laser. As a result of such a high-intensity shockwave, the spherical droplet is transformed into a complex hollow structure which is composed of two spheroidal shells. In our interpretation (see details in Krivokorytov et al., Physical Review E 95, 031101(R) (2017)), the observed evolution of the droplet shape results from cavitation and spallation phenomena that occur after the propagation of the shock wave and rarefaction wave through the body of the droplet.

Laser irradiation of the front surface of the droplet produces a convergent hemispherical shock wave that focuses during the propagation towards the central region of the droplet, causing cavitation. Then the shockwave continues propagating to the rear face of the droplet, where it encounters a free surface. The interaction the shockwave with the rear surface causes the spallation effect. The proposed scenario of the droplet fragmentation is supported by results of numerical simulations.
ELECTRON AND LATTICE DYNAMICS IN MATERIALS WITH LINEAR DIRAC-TYPE DISPERSION STUDIED BY UV - THZ ULTRAFAST LASER SPECTROSCOPY

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We have studied the photoinduced response of the topological insulator Bi₂Se₃ using femto- and picosecond pulses at wavelengths from UV to THz range. Excitation of the crystal by nonresonant mid-infrared laser pulses leads to the formation of electron-hole plasma. We study ultrafast dynamics of this plasma measuring pronounced transient reflectivity signal in a broad range of wavelengths and pump intensities. Ultrafast response of Bi₂Se₃ to intense terahertz pulses is recorded by methods of THz time domain spectroscopy. We observe coherent atomic motions of four various symmetries, the relative amplitudes of which allow evaluation of lattice anharmonicity.

NANOPLASMONIC AMPLIFICATION OF LASER HARMONICS FROM THE INFRARED TO THE MID-INFRARED

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We will present recent results in strong laser field science and nanoplasmonics. Our research is at the frontier of two large and highly successful fields of modern research: ultrafast science and nanoscale physics. On the one hand, ultrafast science produces and analyses the electronic motion at the shortest space (sub-nm) and time scales (attoseconds, with an unprecedented control of the atomic or molecular nonlinear response. On the other hand, nanoscale research opens new routes in extreme optics, from the low to the strong field regime. By bringing together ultrafast laser-matter interactions and nanophysics, we have discovered new routes in boosting the non-linear response using nanostructured photonic crystals. This is a very vast domain of research that encompasses all sorts of nano-objects, as well as meta-materials whose structure can be engineered so as to display some particular electromagnetic properties.

Plasmonics is a very fast growing field which already finds practical applications in numerous aspects of day to day life. An incident laser field, coupled with ad hoc resonant structures, can excite surface plasmons, i.e. charge oscillations at the interface between a metal and a dielectric. By carefully shaping the metallic structure at the nanometre scale, it is possible to manipulate those
oscillations, in order for instance to locally enhance by several orders of magnitude the strength of the electromagnetic field radiated by the oscillation of the charges. While this effect has been applied to multiple nonlinear optics processes (like second harmonic generation, four wave mixing), the extension to HHG has been hampered by two main factors. First the low damage threshold of metallic nanoparticles at IR and mid-IR wavelengths. Second, a difficulty in designing appropriate large amplifying volume resonant structures. Indeed, the bowtie geometry that has been mostly used up to now, while offering a very high localized field enhancement (by up to 3 orders of magnitude), presents nm3 scale volume. We will present two strategies to increase the field enhancement volume by 3 to 6 orders of magnitude. Using our novel “nano-amplifiers”, we have observed the amplification of high harmonic generation from infrared1 and mid-infrared2 laser-crystal interaction by up to 2 orders of magnitude.

Our results open the way towards strong field physics at high repetition rates with low cost small scale femtosecond lasers (oscillators, fibre lasers). Potential applications range from nanoscale imaging, efficient storage and transfer of information to nanoplasmotics (optical filters, waveguides), novel photon and particle sources, and even biomedical sciences.

1. Rana et al, submitted to Scientific Reports
2. Franz et al. submitted to Nature Photonics

PHOTOIONIZATION OF CaF$_2$ SUBJECTED TO ULTRASHORT LASER PULSE IRRADIATION

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We numerically investigate the photoionization of bulk CaF$_2$ subjected to ultrashort laser pulses with duration 220 fs and 515 nm wavelength. At low laser intensities, photoionization is due to multiphoton absorption, while at high laser intensity we find that photoionization is suppressed due to dynamical Stark shift which results in increased bandgap and less ionization. The theoretical result may be helpful in the interpretation of available experimental data on the ablative crater depths produced on CaF$_2$ surfaces by ultrashort intense laser pulse irradiation.
THERMAL MECHANISM OF THZ GENERATION FROM METAL PARTICLES AND SURFACES

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The talk is devoted to the role of ultrafast thermal effects in the process of THz generation from metals and metal structures irradiated by femtosecond laser pulses. Despite the relatively low efficiency, study of optical-to-THz conversion on metal surface are still relevant, mostly because of possible applications in structural diagnostics and due to the fundamental interest in transformation mechanisms. In contrast to research of THz generation during the optical breakdown, there is no clear theoretical interpretation of THz signals emitted by metal surfaces, particles and gratings.

The whole set of experimental data obtained in this field during the last 10-15 years seems difficult to interpret. Duration of the THz signal is almost independent of the metal surface structure and equals to 1-2 picoseconds, which is much longer than the laser pulse duration under the typical experimental conditions. The experimental dependence of the THz radiation energy on the optical fluence varies from quadratic (and slower) to the power laws with the exponent 5-6. It’s also important that the conversion efficiency on metal films strongly depends on their thickness up to the values 100-120 nm (for gold) despite the optical skin-layer is typically 10-15 nm.

We consider thermal mechanism of low-frequency current excitation to interpret the most complicated experimental features of THz radiation. Particularly, in this model THz pulse duration equals to the time of thermal energy relaxation in the initially heated skin-layer, which is about 1-2 picoseconds. Nonlinear energy dependence can be interpreted as a consequence of nonlinear heating when the electron scattering rate depends on the temperature. The possibility of plasmon-enhanced heating and THz generation from metal structures is also discussed.

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ANISOTROPIC THZ GENERATION FROM BISMUTH SURFACE INDUCED BY FEMTOSECOND LASER PULSES

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New experimental results on the terahertz (THz) wave generation from mono- and polycrystalline bismuth samples irradiated by femtosecond laser pulses are presented in the talk. Bismuth was chosen due to its relatively low charge carrier density $5 \times 10^{17}$ cm$^{-3}$ and anisotropic effective mass ranging from $1 \times 10^{-3}$ to $3 \times 10^{-1}$ of free electron mass in vacuum. Therefore, according to previous theoretical studies, the conversion efficiency in bismuth should be higher compared to the case of metals.

The most unexpected result of our observations is pronounceable asymmetry of THz response with respect to half-turn of a monocrystalline sample around its normal. The optical-to-THz conversion efficiency was up to 2 orders higher than for metal at moderate fluence of $\sim 1$ mJ/cm$^2$, while THz energy was proportional to laser pulse energy to power $1.3 \div 1.4$ over the entire measurement range. Also uncommon behavior of THz polarization was observed for polycrystalline samples with parallel strips on the surface. Independently of incident laser pulse polarization the tangential component of generated THz electric field was parallel to strips.

In the case of the polycrystalline sample, the optimal angle of incidence corresponds to the minimal specular reflection of laser light. Similar dependences were observed earlier when THz radiation was generated from metal and metal/semiconductor structures. In those works THz generation was interpreted in terms of some nonlinear effect (e.g., photoelectric) enhanced by the excitation of surface plasmons. In our experiments, optical-to-THz energy conversion was also dramatically enhanced on the structured surface of the Bi sample. In contrast to experiments with metal nanogratings in our case the scale of the strips was much greater than the optical wavelength and they were directed along the plane of incidence in the optimal case. Therefore, we cannot state that the THz generation in the Bi sample is attributed to the surface plasmon excitation. However, some experimental features of optical-to-THz conversion on the Bi surface can be interpreted theoretically in terms of thermal nonlinear response of electrons.

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SPONTANEOUS EMISSION OF ATOMS IN STRONG LIGHT FIELD

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New approach [1,2] to study the spontaneous emission of the atomic system in the presence of the high-intensity laser field is used to study the process of harmonic generation. The analysis is based on the consideration of quantum system interaction with quantized field modes being in vacuum state, while the intense laser field is considered classically beyond the perturbation theory. The numerical analysis of the emission from the single one-electron one dimensional atom irradiated by the femtosecond laser pulse of Ti-Sa laser is discussed. It is demonstrated that not only odd but also even harmonics as well as lines associated with transitions between different discrete levels can be emitted if the laser field is strong enough. The origin of appearance of even harmonics is studied. It is explored that they result from the bremsstrahlung which becomes efficient in the regime of strong ionization. The obtained results are compared with that found in the frames of semiclassical approach widely used to study the harmonic generation. It is found that semiclassical approach is inapplicable in the strong-field limit.

References.


QUANTUM-MECHANICAL EFFECTS IN EXCITATION OF LOW-FREQUENCY CURRENTS DURING GAS IONIZATION BY TWO-COLOR LASER PULSES
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We examine a new method for generation of the coherent few-cycle mid-infrared (MIR) pulses. The method utilizes the gas ionization by ultrashort incommensurate two-color laser pulses, which contain the fields at two different frequencies. One of the frequencies is detuned from the doubled value of the other one. Such incommensurate pulses can be obtained with the use of the secondharmonic generation crystal or optical parametric amplifier [1-3]. In previous works the radiating low-frequency currents excited by such pulses were calculated analytically and numerically on the basis of semiclassical approach [1-4]. It was shown that excitation of radiating mid-infrared electron currents can be understood as the ionization-induced wavemixing caused by response of free electrons [4]. The duration of excited low-frequency current is much less than the laser pulse duration, and the central frequency can be controlled by tuning the frequencies of the optical field components.

In this work we calculate the electron current excited by the two-color pulse in a gas through the use of the analytical and numerical solution of 3D time-dependent Schrödinger equation (TDSE). The dependences of the duration and energy of the generated MIR pulses on the parameters of the laser pulse are found. It is shown that the results of numerical quantummechanical calculations are in good agreement with the results given by the semiclassical approach at high intensities corresponding to the tunneling ionization regime. At lower intensities, on the contrary, qualitative differences are observed in the dependences of the duration and energy of MIR pulses on the intensity and wavelength of the main field. According to the quantum-mechanical approach, the pulse duration makes large oscillations, and the energy of the pulses makes jumps at certain values of intensity and wavelength. On the basis of the analysis of the electron velocity distribution function, it is shown that above-mentioned effects are associated with the opening and closing of ionization channels of an atom during the change in the intensity or wavelength of the main field of the laser pulse.

FREQUENCY UP-SHIFT IN THE STIMULATED THERMAL SCATTERING IN TWO-PHOTON ABSORBING COLLOIDS OF METAL NANOPARTICLES

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Stimulated scattering is one of the most efficient physical mechanism to generate frequency shifted coherent light emission and optical phase-conjugate waves. Among other scattering mechanisms, the stimulated thermal scattering (STS) is sufficiently rare-studied nonlinear-optics effect since 60-th, which in part is due to significant technical difficulties in observation. Recent renewal of the interest in STS is related to studies in multi-photon absorption spectroscopy [1]. Experimental study of STS in Ag nanoparticle colloids has been recently reported [2].

In [1], STS was studied in counter-propagating two-pump wave scheme in two-photon absorbing dye solution. A number of quite unusual features is reported, namely the absence of frequency shift and extremely low threshold value, which are in contrast to the most of conventional stimulated scattering mechanisms.

Here, we report the results of our study of main features of STS in two-photon absorption in colloids of metal nanoparticles. We consider the four-wave mixing geometry in which two pump waves of the same frequency propagates counter to each other. Counter-propagating Stokes and anti-Stokes signals are amplified as a result of STS process, which is treated self-consistently using wave equations for the Stokes and anti-Stokes signals coupled through the perturbation of temperature which is found using solution of the thermal conductivity equation. Amplification of signal waves proceeds not only through the scattering on the spatial thermal grating which is formed by counter-propagating pump waves, but through interaction with spatially uniform coherent temperature oscillations which are of equal phase in all the interaction region. Interaction results in formation of self-induced cavity which makes it possible for lasing in this system. We find that in contrast to results of Ref.[1], four-wave mixing two-photon absorption STS scheme is characterized by non-zero frequency shift (up-shift or down-shift, depending on the sign of temperature derivative of the dielectric permittivity of nanoparticle’s colloid).
The value of frequency shift depends on the power ratio $\alpha$ of counter-propagating pump waves and has the minimum value of $\sim 0.5\lambda T q^2 / \rho_0 C_v$, $\lambda T$ is the heat conduction coefficient, $q$ is the characteristic wave number of the spatial temperature grating, $C_v$ is the specific heat at constant volume, and $\rho_0$ is the unperturbed density.

We report the results of our experimental measurements of the frequency shift in STS in two-photon absorbing colloids of metal nanoparticles which are in perfect agreement with our theoretical results.


**GRAPHENE IN STRONG ELECTROMAGNETIC FIELDS**

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Since the synthesis of graphene in 2004, its investigations have been attracting tremendous interest due to its unique physical properties and numerous potential applications. Hundreds of publications devoted to graphene appeared, including several reviews. In our presentation, we will discuss some specific nonlinear properties of graphene revealed in strong electromagnetic fields.

In particular, in the first part of the presentation the experimental and theoretical results on optical emission from graphene irradiated by a strong (up to 300 kV/cm) short terahertz pulse will be presented. In experiment, the appearance of emission from graphene was observed in the 350-600 nm range at high THz fields. We detected the increase of the optical emission by nearly 3 orders of magnitude while the THz field increased by a factor of 2 only. The spectrum of the optical emission corresponded to Planck’s law for the spectral radiance of a black (grey) body. We
attribute the optical emission to bias-induced spontaneous emission from energetic charge carriers in the graphene. On the base of the theoretical analysis, we revealed the importance of dynamic (ballistic) mechanism of electron-hole pair generation induced by intense terahertz pulse with subsequent spontaneous radiative recombination.

In the second part of the presentation we will discuss our results on the generation of the second optical harmonic (SHG) of laser radiation from graphene in the presence of a high power terahertz pulse. We demonstrated essential amplification of SHG under the action of THz field as compared to its absence. Polarization characteristics of SHG specific for graphene will be presented together with dependences of SHG efficiency on optical and THz fields. Detailed theoretical analysis of von Neumann equation allowed us to propose a new mechanism for SHG in graphene under the combined action of optical and THz fields. This mechanism is caused by broadening of the interband resonance due to quasi-particle acceleration by an intense THz field in the process of interband transition. As a result of the “field” interband resonance broadening, the region of electron-hole generation in k-space becomes asymmetric, which leads to the appearance of necessary anisotropy allowing for SHG in dipole approximation.

ELECTRON MODES OF COLD PLASMA FORMED AT ATOM PHOTOIONIZATION BY A SHORT LASER PULSE OF CIRCULARLY POLARIZED RADIATION

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When the atoms of matter are ionized by a field of intense laser radiation, non-equilibrium plasma with an anisotropic photoelectron velocity distribution is formed. The difference in distribution of photoelectrons from the Maxwell distribution leads to the fact that many properties of photoionized plasma differ qualitatively from the properties of weakly non-equilibrium plasma.

In this report, the dispersion properties of the electron eigenmodes of plasma formed as a result of the photoionization of the matter atoms by a short laser pulse of circularly polarized radiation are investigated. The peculiarity of such plasma is strong anisotropy of the photoelectron distribution, which is associated with large electron kinetic energy in the plane across the anisotropy axis determined by the direction of the ionizing pulse propagation. The analysis was carried out by us in the framework of the cold plasma model, which does not take into account the thermal spread over photoelectron velocities. This approach allowed us to obtain the main qualitatively new features of the electron perturbation dispersion in such plasma in a simple analytical form.
In the region of frequencies exceeding the electron Langmuir frequency there are two branches of electron waves in photoionized plasma. The dispersion law of the wave branch, whose electromagnetic field is close to the transverse field and the phase velocity is close to the speed of light, slightly differs from the analogous law of electromagnetic wave propagating in a Maxwellian plasma with the same electron density. On the contrary, the dispersion law of another high-frequency wave branch, which characterized by small phase velocities in comparison with the speed of light and practically longitudinal electric field, is new and qualitatively differs from their analog in isotropic Maxwellian plasma - Langmuir waves. The dispersion of the waves considered by us is the stronger, the greater the angle between the direction of their propagation and the anisotropy axis of the photoelectron distribution. The frequency of the considered waves in the photoionized plasma can exceed the electron Langmuir plasma by several times.

Also it is shown that in the range of the frequencies much smaller than the electron Langmuir frequency the development of aperiodically unstable perturbations whose electric field can be either purely transverse or longitudinally-transverse is possible in photoionized plasma. The anisotropic dependencies of the increments of these modes on their wave vector are obtained and the instability regions of these modes in the space of wave vectors are found. The ratio of the maximum instability increment of both modes to the electron Langmuir frequency is determined by the small ratio of characteristic transverse electron motion velocity to the speed of light.
OPTICAL PROPERTIES OF METALS STIPULATED BY ELECTRON-PHONON INTERACTION

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Optical properties of metals at moderate temperatures (lower than Fermi ones), stipulated by electron-phonon interaction, are described using dielectric function (DF). The expression for DF is derived using quantum statistical approach and linear response theory, in a single-moment longwavelength approximation, which results in Drude-type expression for DF with complex effective collision frequency. The common expression for effective collision frequency of electrons for the case of electron-phonon interaction is derived using the Frohlich Hamiltonian and the first Born approximation. On the base of this expression simple formula for effective collision frequency is obtained for the case of transitions in single zone and with single phonon mode. This formula makes one possible to take into account different temperatures of electron and ion systems for the case of metals heated by short (femtosecond) laser radiation and is valid in wide range of radiation frequencies, from infrared to X-ray. The generalizations of this approach to the cases of intraband transitions and Umklapp scattering are discussed. The proposed approach opens possibility to spread the derived early wider range (on radiation frequencies) model of DF, valid for the case of temperatures higher than Fermi one [1], to the case of lower temperatures.

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The nonlinear wave mixing and associated parametric frequency conversion present one of the most successful ways for generation of ultrashort electromagnetic pulses in infrared, optical, and ultraviolet bands (including generation of extremely short few-cycle electromagnetic pulses). Such generation usually utilizes nonlinear crystals as a working medium where the low-order wave mixing (three- or four-wave mixing) can take place. In this work, we study the atypical multiwave (high-order) mixing that accompanies ionization of a medium in a strong field of the ultrashort laser pulse and show how this wave mixing may result in generation of few-cycle electromagnetic pulses in infrared, optical, and ultraviolet bands. When the generation of extremely short few-cycle pulses is considered, the method that is proposed here and is based on the ionization-induced multiwave mixing has a number of advantages over the parametric conversion employing nonlinear crystals.

Some advantages originate directly from the high-order nature of the strong-field ionization: the large number of mixed waves causes the duration of the generated pulse to be much shorter than the duration of the ionizing laser pulse so that the direct generation of the much shorter pulses is possible (and there may be no need for external compression or other additional efforts for generation of few-cycle pulses). We study the most basic configuration for ionization-induced wave mixing where a two- or three-color ionizing pulse is just focused in a gas. We calculate analytically and numerically the free-electron currents excited in a formed plasma at combination frequencies of the ionizing pulse and the electromagnetic radiation generated by these currents. The analytical and numerical results show that the duration of the generated pulse is equal to the ionization duration and may be close to the sub-cycle limit for the generation in the mid-infrared range and may be as short as of 1–2 fs in the ultraviolet range (when three-color pulses are employed). The estimations for the radiated energy show that the conversion efficiency can be as high as a few percent (for the mid-infrared band) even in this basic configuration when no efforts are made to improve synchronism conditions and the propagation distance. The obtained analytical formulas describe the main scale dependences of the properties (amplitude, duration, carrier-envelope phase, and spectral shape) of generated pulses on the parameters of the ionizing laser pulse (intensities of the one-color components, duration, and phase structure) and reveal the similarities and dissimilarities between the ionization-induced wavemixing under consideration
and the low-order wavemixing in a common nonlinear media. This may prove useful for the identification of the ionization-induced wavemixing in the experiments as well as for designing and optimizing schemes for efficient generation of the few-cycle pulses.

**ON INTERACTION OF H-WAVE WITH METALLIC – DIELECTRIC PHOTONIC CRYSTAL**

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An interaction of electromagnetic H-wave with one-dimensional metallic – dielectric photonic crystal consisting of metallic and dielectric alternating layers, is studied numerically. The reflectance, transmittance and absorptance power coefficients are considered. One detects the frequency zones of the power coefficients behavior. It is shown also that the quantum wave and spatial dispersion effects of electron plasma contribute to the power coefficients in the places of their sharp behavior.
EFFECTS OF Fs LASER BEAM ON (Ti/Al) NANO-LAYER THIN FILM

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The effects of ultra-short laser pulses on reactive titanium and aluminum (Ti/Al) nano-layer thin film (NLTFT) were investigated. The NLTFT, composed of alternated Ti and Al nano-layers, was deposited by ion-sputtering. The thickness of multilayer structure was 200 nm. It consisted of five Ti/Al bilayers and thickness of particular Ti and Al layer was 17 nm. The NLTFT surface was finished by Ti layer thickness of 27 nm. Single pulse irradiations were conducted in air with focused and linearly polarized laser beam- 515 nm wavelengths and 200 fs pulse duration. Laser beam was Gaussian with 6.7 μm 1/e- beam diameter. In the experiment pulse energy was from 0.02 up to 1.30 μJ. We investigated how increase of pulse energy affected the morphological changes of the irradiated nano-layer thin film. Effects of laser induced composition and morphological changes were monitored by different microscopy techniques and profilometry. Following results were obtained: (i) one step partial/selective ablation of upper Ti layer from nano-layer Ti/Al at low laser pulse energies and (ii) ablation of complete nano-layer Ti/Al at higher. Single pulse selective ablation of the upper Ti layer was confirmed based on profilometry along the ablation steps and reduction of Ti concentration in the ablated areas. Profilometry has revealed that action of single 200 fs pulse with pulse energy/fluence of 0.08 μJ/320 mJcm⁻², as well as 0.16 μJ/640 mJcm⁻², were able to ablate the first Ti layer from the nano-layer structure without creating significant modification of the layers beneath. One very important parameter for laser processing such as ablation threshold fluence was found to be Fab = 88 mJcm⁻². The results can be useful in laser processing of reactive Ti-Al thin film without considerable unwanted heat released effects which could be occur because of negative enthalpy of mixing of the constituents. In the present study, to our best knowledge, for the first time selective ablation of the Ti top layer from the nano-layer metal-metal thin film structure was reported. Also, ablation threshold for used laser and NLTFT was calculated for the first time.
References:


SINGLE-SHOT INTERFERENCE STRUCTURING OF Cu2O FILMS BY PICOSECOND LASER PULSES

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The laser modification with the following metal particles appearance of thin oxide metallic films is a promising method for the formation of binary metal structures on the glass substrates. That find different application in photonics and plasmonic. It is important to confirm that even a single ultrashort laser pulse irradiation is able to form the metal layer that makes possible to imprint structures with required shape.

Formation of the interference pattern was achieved in the confocal scheme, where phase grating was applied as a laser beam splitter (fig. 1 «a»). Phase grating was fabricated by laser-induced μ-plasma technology [1] and pre-calculated to achieve high diffraction efficiency. The beams were overlapping beneath the second lens, creating an interference pattern in their intersection area. The diaphragm set between the lenses to block the zero- and higher orders of the diffracted beams. As a light source we used Nd:YAG laser (operated at 355/532/1064 nm wavelength) with 25 ps pulse duration. High compact structures (with period ~ 0.5–1 μm) were achieved as a result of laser interference micro-patterning of CuOx film (fig. 1 «b»).
The period of the stripes is determined by the radiation wavelength \( \lambda \) used for its formation and angle \( \theta \) between converging beams. The numerical aperture of the focusing microobjective limits the period of the formed structures in the scheme. One way to increase the processing field is to use the multiplexing of interference pattern when moving the sample relative to the laser beam. The formation of a large field was carried out by a discrete displacement of the coordinate table after the formation of an interference pattern from the formation of a single laser pulse. Fig. 1 «c» shows the result of such microprocessing with field multiplexing.

![Figure 1. The scheme of laser interference micro-patterning (a); results of two-beam interference patterning of CuOx film: single shot (b), field multiplexing (c).](image)

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RELATIVISTIC PLASMA PHYSICS WITH TABLE TOP TW FEMTOSECOND LASERS: SCIENCE & APPLICATIONS

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We present results of the experimental and numerical studies of hot electron generation and high efficient gamma production by the interaction of relativistically intense laser pulses with solids.

Special attention was paid to optimization of the target and laser radiation parameters. We used the Ti:Sa laser system (p-pol, 800 nm, 10 Hz, 40 mJ, 45 ± 5 fs and I_{max} = 5×10^{18} W/cm^2, ASE & prepulses <10^{-8}). For some experiments contrast rations at the nanosecond and picosecond time scales were adjusted to achieve the desired phenomena. In other experiments the Nd:YAG laser (1064 or 532 nm, 30-120 mJ, 6 ns, I~10^{12} W/cm^2) was used to create the controlled long and dense pre-plasma layer. This laser was locked with the Ti:Sa laser system with accuracy better than 1 ns. Different targets were used: metal plates made of Fe, Mo, W, thick polymer films and plates, as well as different structured targets. We also discussed applications of our studies in nuclear physics, microelectronics, and other areas of research.

PLASMONIC NEAR-FIELDS ABLATION ON SI INDUCED WITH AN INTENSE FEMTOSECOND LASER PULSE

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The excitation of the surface plasmon polaritons (SPPs) at the interface between non-metallic materials has been proposed as one of dominant physical mechanisms for the formation of a periodic nanostructure on solid surfaces through ablation induced with the intense femtosecond (fs) laser pulses [1]. This phenomenon has attracted considerable interest since the first observations [2], because the selforganized nanostructure suggests a potential route to transcend the diffraction limit in the laser-matter interactions. By observing anomalous optical absorption of a Si grating, similar to the so-called Wood’s anomalies [3], we present clear evidence that SPPs can be resonantly excited at a Si–air interface with an intense fs laser pulse for the first time to our knowledge.
As shown in Fig. 1(a), we prepared the laminar Si gratings fabricated by photolithography and a dry-etching process. Polished $p$-type crystalline Si (100) was used as the substrate. We measured the reflectivity $R$ at the grating surface by using a linearly polarized, 800 nm, 100 fs laser pulse from a Ti:sapphire laser amplifier system operated at a repetition rate of 10 Hz. The fs pulse was focused on the Si grating surface in air with a lens at an incident angle $\theta$ ranging from $10^\circ$ to $40^\circ$. The reflected pulse was expanded by a pair of lenses. The spatial intensity distribution was observed with a charge-coupled-device camera to evaluate $R$ in the central beam area.

For a comparative study, we made the experiment at two different laser fluences. One was a low fluence of $F_1 = 40 \text{ mJ/cm}^2$ at which the fs pulse never excited high-density electrons in the Si surface. The other was a high fluence of $F_2 = 700 \text{ mJ/cm}^2$ much above the single-shot ablation threshold of the Si surface for exciting high-density electrons, leading to formation of a metal-like layer on Si. In the experiment at $F_2$, we translated the Si target in the direction parallel to the surface shot by shot so that the laser pulse hit the fresh surface area. Figure 1(b) shows $R$ for the Si grating measured with a single shot of the fs pulse as a function of $\theta$. At $F_1$, $R$ for both polarizations changes monotonously with increases in $\theta$. At $F_2$, $R$ for the $p$ polarization decreases distinctly for $\theta$ from $23^\circ$ to $25^\circ$, namely displays a dip, while the dip is not seen in $R$ for the $s$ polarization. The appearance of the dip resembles the anomalous optical absorption appearing in reflection at a metal grating surface observed by Wood [3]. Calculation for a model target reproduces well the experimental results of the reflectivity. The experimental results obtained clearly show that SPPs at a Si–air interface can be resonantly excited with an intense fs laser pulse.

We also observed the surface of the irradiated target with a scanning probe microscope. Using the $p$-polarized laser pulse at $\theta = 24^\circ$, the nanogrooves of 400 nm in depth was observed to be clearly formed on the surface through the laser ablation. The experimental results demonstrate that intense periodically enhanced near-fields of SPPs ablate the target surface to form the laser-induced periodic surface structure.
Fig. 1 (a) SPM image of the Si grating surface. (b) Reflectivity of the Si grating measured with a single fs laser pulse at $F_1 = 40 \text{ mJ/cm}^2$ (blue) and $F_2 = 700 \text{ mJ/cm}^2$ (red) for $p$ (solid) and $s$ (open) polarizations as a function of the incident angle. The lines connect the points to support visualization.

References


Ultra short laser radiation in the fsec time domain (8-several 100 fsec) represents nowadays a fascinating and challenging tool for modifying and structuring surfaces. One of the particularly interesting aspects definitely arises from the fact that fsec light pulses exhibit a very wide range of pulse shapes (in the time domain) as well as an intrinsically complicated frequency spectrum. As a consequence, many different physical processes can and will result by changing the laser parameters. In an ideal situation, these parameters can be used to control the outcome of the laser material modification process and drive it to optimized results. However, as it seems it still a long way to go to understand all the different processes and their dependence on laser parameters before a reliable tuning of an optimized surface modification will be possible.

In this overview, we will report on recent attempts to reach this goal by experiments, which have the goal to correlate the particle emission (laser ablation) and the development of surface periodic structures (laser induced periodic surface structures, LIPSS) under various laser parameters (fluence, pulse length, wavelength, number of pulses etc.).

The mass spectrum of emitted particles, their energy distributions, thresholds for ablation and the ion to neutral yield have been studied.

In order to verify the possibility of the occurrence of ultrafast electronic ablation process in metals (Al, Cu, Ti, Au and Ag) exposed to ultrashort laser pulses velocity distribution, pulse duration and pulse energy dependence studies for both ablating and post-ionizing pulses were used. For all samples three distinct different velocity groups were observed. For instance, velocity distribution of Ti shows three peaks at 7, 3 and 0.3 ev. The first two peaks indicate the presence of fast particles which can be created via ultrafast electronic processes such as Coulomb explosion and rapid plasma formation. The third peak is the signature of thermal process.

This allows to identify different processes, characteristic for fsec laser pulses interacting with surfaces. These experiments have to be performed in UHV. Consequently, the development of LIPSS structures and, in particular, so-called high frequency and low frequency structures (HSFL and LSFL) have been investigated under UHV conditions.
Various possibilities to simulate the development of surface structures will be discussed. Taking into account the wealth of physical processes and the number of atoms involved is a challenging task.

**Figure 1**: Neutral signal of Ti atoms as a function of the time of flight (TOF) between target and post-ionizing laser beam. For better visualization, the energy spectra is presented here in this form. However, the TOF spectra can be directly converted into a velocity or energy distribution.
I will report on our progress in the development of 3D laser printing technologies for fabrication of microoptical components [1,2] and complex nanoparticle structures [3]. Fabrication, characterization, and applications of the generated microoptical components and nanoparticle arrays in nanophotonics, plasmonics, and optical sensing will be demonstrated and discussed.

Fig. 1 Experimental setup applied for laser printing of nanoparticles

References
PROCESSING OF TRANSPARENT DIELECTRICS WITH ULTRAFAST BESSEL BEAMS: FROM HIGH ASPECT RATIO VOID FORMATION TO STEALTH NANOMACHINING


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Ultrafast lasers have revolutionized materials science because of the high peak power they can generate even in the bulk of transparent materials. They allow for extremely localized nonlinear ionization. Besides, the processing of glass and sapphire is of recent particular importance for applications in consumer electronics (displays, touch-screens), microelectronics, substrates for LED growth, etc. However, conventional ablative techniques are slow and generate a large amount of debris. In contrast, stealth dicing enables material separation with minimal debris [1]. It consists of creating an internal damage defining a plane, which leads to a fracture under mechanical bending or stretching. This eventually leads to material separation at high illumination speed.

In this context, we show that ultrafast Bessel beams are very advantageous. We have demonstrated that they allow controlling quasi-uniform energy deposition within transparent dielectrics. This is highly differs from the filamentation process created by Gaussian beams which undergo nonlinear distortions [2]. In single shot, Bessel beam generate formation of high aspect ratio nanochannels appearing either as a through-channel, or as a terminated channel opened on only one of the sample's surfaces [3] or as voids fully enclosed within the material's bulk [4]. The last case shows that the pressure created after picosecond or femtosecond pulse illumination is enough to compress the material around the void. We have shown the high influence of the pulse duration on void morphology.

Creating channels side by side allows for inducing a weakened plane for the stealth dicing process. We report novel results where we engineer the transverse intensity profile to control the tensile stress distribution within the material and allow for a precisely defined cleaving plane. We show that the high precision of the fracture plane with regard to the laser-produced void is a particularly important parameter for the strength of the material after the separation. Finally, we report a new and different approach for crystal cleaving where the cleaving plane is created by controlled crack formation [5].

The research leading to these results has received funding from the European Union FP7 programme (GA 619177-TiSaTD), European Research Council (ERC) under the European Union's Horizon 2020 programme (GA 682032-PULSAR), from the Region Franche-Comte. This
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FEMTOSECOND-PULSE INSCRIPTION OF PHASE-SHIFTED FBGS AND ITS APPLICATIONS.
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Phase-shifted fiber Bragg gratings (PSFBGs) are widely used in distributed-feedback fiber (DFB) lasers, optical sensors for simultaneous temperature/stain measurements, high-sensitivity ultrasonic detectors, optical signal processing and filtering elements. Traditionally, PSFBGs are inscribed by UV radiation via a phase mask with a phase shift in the structure, or by means of a uniform phase mask displacement in the process of inscription, or by a postprocessing of a uniform FBG.

The alternative way of FBGs inscription is a rapidly growing technique of femtosecond (fs) laser modification of refractive index (RI) in transparent materials. The key advantages of this technique are possibility of RI change of non-photosensitive materials, inscription of FBGs through the fiber plastic coating, which is transparent for IR fs radiation, and possibility of 3-D structure formation inside material that enables FBGs inscription by direct point-by-point writing technique without a phase mask.

The results of long high-quality PSFBGs inscription in passive and active fibers by point-by-point technique using fs laser pulses are presented. Phase shifts are introduced during the inscription process with a piezoelectric actuator, which quickly shifts the fiber along the direction of its movement in a chosen point of the grating with a chosen shift value. The developed technique allows us to inscribe both single- and multiple PSFBGs. The maximum PSFBG length achieved
in the experiments is 34 mm in passive fibers, which provide corresponding transmission peaks with bandwidth less than 1 pm that is the smallest value obtained by fs direct point-by-point writing technique.

The generation single-frequency radiation at 1550 nm with bandwidth of 20 kHz and signal-to-noise ratio of >70 dB is shown by using 37 mm π-phase-shifted FBG inscribed in an active Er-doped fiber and forming DFB laser cavity.

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FORMATION OF TRANSPARENT THERMOCHEMICAL LIPSS ON THIN CR FILMS BY FEMTOSECOND LASER BEAM SCANNING
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Laser micromachining of materials is intensively developed in the last decade in order to change or to provide new features for their surface. From practical point of view, cost-effective technologies of the surface modification with the large area are required. The formation of laser-induced periodic surface structures (LIPSS) appears to be useful for that, since it is based on the self-ordering mechanism when laser radiation is focused in the spot size that is much larger than period of the structures. The formation of new type of LIPSS – thermochemical one (TLIPSS), arising not from ablation, but from surface oxidation of thin metal film was recently demonstrated. In this case, the surface structure grows up without formation of debris formed by redeposition of ablation products.

The experimental results on the formation of the TLIPSS on the chromium films (deposited on glass substrates) with a thickness of about 20-30 nm at different experimental conditions are presented in this paper. The behavior of processing rate of TLIPSS formation for the thin films differs from one for thick (400 nm) Cr films. The TLIPSS uniformity in case of thin film significantly depends on processing rate with high uniformity up to 18 µm/s at 15 mW and up to 30 µm/s at 21 mW. The period of TLIPSS does not depends on the pulse power and equals to 690 nm at 1 µm/s. At the same time, the period of TLIPSS significantly depends on the films thickness and increase from 690 nm to 950 nm with an increase the film thickness from 25 to 400 nm. This
dependence of TLIPSS period is explained in terms of plasmons theory of TLIPSS formation, the
calculation based on it provides values that is close to experimental one.

After laser processing the TLIPPS structures formed on thin Cr films were etched the selective
liquid etchant containing 6 parts 25% aqueous solution of K3Fe(CN)6 and one portion of 25%
aqueous NaOH solution (the etchant is used in microelectronics industry). Cr oxide layer has much
slower etching rate in the solution than metal film. Completely transparent oxide grating has been
formed after the etching. It can be used for example as diffractive grating with low diffraction
efficiency or as mask for dry etching of the substrate (Cr oxide has much smaller etching rate in
RIE ICP process than pure Cr film at usage of chlorine-containing gas).

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LASER-ASSISTED IMPRINTING OF SURFACE GRATINGS ON A GOLD FILM.
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The processes of laser-assisted imprinting of regular surface relief gratings are experimentally
and theoretically investigated. These processes include recording a spherical nanoantenna on gold
film surface by the first laser pulse and imprinting the grating by the second laser pulse due to
constructive interference of nanoantenna’s radiated surface plasmon waves with the second
incident pulse’s tail. As a first step towards understanding complex phenomena involved in the
formation of such a grating we analytically calculated the distribution of absorbed electrodynamic
power in the gold film and showed that this distribution corresponds closely to the grating profile.

GIANT DOUBLE-RESONANCE ENHANCEMENT OF STIMULATED LOW-
FREQUENCY RAMAN SCATTERING IN HYBRID METAL-CAPPED
NANODIAMONDS
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Magnetic dipolar Mie-resonance of nanodiamonds enables their highly-efficient stimulated
low-frequency Raman scattering via 20-ns ruby laser excitation of their fundamental breathing
mode, with strong additional plasmonic enhancement of the Raman conversion efficiency upon
ablative capping of the resonant nanodiamond core by a silver nanoshell with a broad overlapping
electrical dipolar Mie-resonance.
FABRICATION OF IR-SENSORS BY FEMTOSECOND LASER PULSES

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ABSTRACT

As a sample, in our experiments we used a 30 nm thick silver (Ag) and a 60 nm thick Au/Pd film with an optical-quality surface, deposited onto a 1 mm thick CaF2 substrate by magnetron sputtering. The film was arranged on a 3D motorized micro-stage under PC control. The microholes were produced via single-shot ablation of the film by moderately (NA ≈ 0.25) focused 515 nm, 220 fs TEM00-mode laser pulses. Diffraction grating with diameter $D \approx 3\,\mu m$, period $P \approx 6\,\mu m$ and diameter $D \approx 4\,\mu m$, period $P \approx 6\,\mu m$ of microholes in the gold and silver film formed by strongly focused ultrashort laser pulses with the energy density $F = 0.5\,J/cm^2$ and $F = 0.27\,J/cm^2$, respectively. The resulting sensors were characterized by means of scanning electron microscopy (SEM, JEOL 7001F). IR unpolarized transmittance of the substrate, film and the sensors (both—clean and with an effective, on average, monolayer of rhodamine 6G molecules deposited from its 100 pM ethanol solution by a drop-drying technique, was measured in vacuum in the near-mid IR range of spectral wavenumbers $\nu = 400–5000\,cm^{-1}$, using a FT-IR spectrometer V-70 (Bruker).

A two dimensional square transmission diffraction grating consisting of micron holes in a gold film exhibits surface enhanced infrared absorption of rhodamine 6G in the range of 1400–1600 cm$^{-1}$ near the edge of the lower allowed band with a gain of about 10 (Fig. 1).

Selective IR absorption at 1261 cm$^{-1}$ enhanced by 455 times, was demonstrated for rhodamine 6G molecules, covering a 2D-photonic crystal, represented by a regular array in a 30 nm thick silver film. The reference absorption lines were taken near 2900 cm$^{-1}$, where the IR radiation is freely channeling through the microholes, indicating the reference substrate coverage by the dye molecules for its relative internal calibration. The limit of background-free detection for the analyte was determined at the level $\sim 10^{-2}$ monolayer (Fig. 2).
Fig. 1. a) Scanning electron microscopy image of the transmission diffraction grating (diameter $D \approx 3 \, \mu m$ and period $P \approx 6 \, \mu m$) of microholes in the gold film formed by strongly focused ultrashort laser pulses with the energy density $F = 0.5 \, J/cm^2$ in the single pulse ablation regime. b) (Color online) Infrared spectra of the transmittance $T$ of the dye layer on (lower curve) diffraction grating and (upper curve) CaF$_2$ substrate. The inset shows the magnified image of the low-frequency spectrum of the dye on the substrate.

Fig. 2. a) SEM top view of the microhole array with $D \approx 4 \, \mu m$ and the magnified view of such a single microhole (inset). b) IR transmission spectra of the CaF2 substrate with the R6G monolayer (black curve), and the large-hole sensor with (blue ‘R6G’ curve) and without (purple ‘clean’ curve) such monolayer. The insets with the related highlighting red circles indicate minor R6G absorbance bands used for the near-IR internal calibration and evaluation of enhancement. The red arrows point to the SEIRA-enhanced R6G absorption bands in the mid-IR range (the strongest one at 1261 cm$^{-1}$).

Using a micro-hole grating in a supported silver film as a laser-fabricated novel optical platform for surface-enhanced IR absorption/reflection spectroscopy, characteristic absorption bands of *Staphylococcus aureus*, especially – its buried carotenoid fragments – were detected in FT-IR
spectra with 10-fold analytical enhancement, paving the way to spectral express-identification of the pathogenic microorganisms (Fig. 3).

**REFERENCES**


DIRECT LASER PRINTING OF PLASMONIC NANOSTRUCTURES AND THEIR APPLICATION.

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A simple and high-performance laser-assisted technique for plasmonic nanostructures printing, which includes the ablation of noble metal film (Au, Ag, Au/Pd or Cu) with focused nano- or femtosecond laser pulses followed by processing with accelerated ion beams or plasmon-polariton waves. Plasmonic elements including nanoscale cupolas, voids, jets, rings, holes, crowns, chiral nanojets, porous cupolas and rings and periodic structures can be fabricated on both metallic and dielectric surfaces. The geometry of these nanostructures was shown to be well tuned over a wide range varying both excitation conditions (focal spot diameter, orbital angular momentum, pulse duration and energy) and metal film characteristics (sputtering conditions, thickness, chemical composition). Underlying physical mechanisms responsible for the formation on different functional plasmonic nanostructures are revealed and discussed. All the structures possess pronounced and well-tuned geometry- and size-dependent plasmonic response in visible spectral range, which can be utilized to enhance the photoluminescence or surface-enhanced Raman scattering signals from organic dye molecules and rare-earth complexes.

STRUCTURED LASER BEAMS: SHAPING OF LIGHT ON A FEMTOSECOND TIME SCALE

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Structured laser beams is a term, which is often used for describing light fields with a certain configuration of complex amplitude or polarization distribution. In this work, we elucidate the features of time behavior of focused structured laser beams. By adding the time dimension, one can introduce, similar to the term “(spatially) structured light”, an idiom “space-time structured light”. We demonstrate explicitly the phase delay between transverse and longitudinal components of the light field generated at the focus, i.e. their appearance/reaching the peak at different instances of the optical period, for linear or radial incident polarizations. For other incident
polarization states (circular, circular with a + or – first order vortex, radial with a vortex) the longitudinal field component coexists simultaneously with one or both x and y field components.

Moreover, we report on a remarkable property of cylindrically polarized light beams dressed with/containing a vortex/orbital angular momentum: when sharply focused, azimuthally (radially) polarized light beam containing a vortex generates subwavelength structures such as an electric (magnetic) dipole or propeller/stirrer which rotate at optical frequencies. Ultrafast rotating dipole produced by focusing an azimuthally polarized vortex beam differs significantly from a pattern obtained by focusing circularly polarized light. The numerically calculated field components distributions are verified by simplifying the system with an application of a narrow ring aperture allowing to obtain precise analytical expressions confirming the phase delay between different field components. This tailoring of local optical fields in a subwavelength domain can be used in ultrafast modification/modulation of physical properties of media, e.g. control of electrical or optical conductance between two-terminal junction.

**STRUCTURED LIGHT GENERATED BY DIFFRACTIVE OPTICAL ELEMENTS FOR LASER NANOFABRICATION**

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In the last decade so-called structured laser beams spread out wide in laser processing of materials, optical and quantum communications, optical microscopy and biophotonics. The possibility to manage not only the distribution of the formed laser beam intensity, but also its phase distribution and polarization is critically important when performing laser nanostructuring to create so-called metacoatings or metasurfaces. The control of laser characteristics, such as amplitude and phase distribution in the beam and its polarization state, during the procedure of structuring allows to form plasmonic structures with radically differing topologies. Diffraction optics allows to realize such control via single diffractive optical elements and their combinations.
Diffraction elements that form structured laser beams, can be used not only on the stage of laser fabrication of metasurfaces and metacoatings, but also during their analysis and characterization. Also, there is an opportunity to tune the structure of a light field formed in such a way by the means of the size mismatch between diffraction element and the irradiated laser beam.

At the moment diffractive optical elements are widely used both for the formation of specified complex field distributions, including so-called vortex laser beams carrying OAM. Besides they are used to realize polarization conversion and also for forming fields with a longitudinal field component. Damaging threshold of such optical elements is much higher because of low material absorption of the substrate, on which they are fabricated. One more advantage of diffractive optical elements is worth mentioning – their efficiency (diffractive optical elements allow theoretically to modulate falling light with an efficiency of up to 100 %). So, laser nanofabrication using structured laser beams formed via diffractive optics elements gives an opportunity of developing methods of high-speed and efficient laser nanofabrication of functional sensor elements with variable morphology for different states of laser pump polarization.

In this work, we present various methods of design diffractive optical elements forming structured laser beams and some results demonstrating possibilities of using such elements for ultrafast laser nanofabrication.
ELECTRON-PHONON-MAGNON INTERACTIONS OPEN THE DOOR TO FEMTOSECOND LASER NANOSTRUCTURING

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Hybrid metal-ferromagnet multilayer structures play an important role in nanophotonic applications [1,2]. Here we show how ultrafast pump-probe measurements on hybrid metal-ferromagnet bilayer structures can help predict a novel physical phenomenon: ultrafast transient overheating of ferromagnetic layer excited by hot electrons through a macroscopically thick layer of noble metal. Combined with the numerical two-temperature model simulations of ultrafast heat diffusion, our experimental observations reveal the dominant role of the interfacial thermal boundary (Kapitza) resistance at metal-ferromagnet interfaces as well and a difference in the electron-phonon coupling strength between noble and ferromagnetic transition metals. Excitation of exchange-coupled magnons by non-equilibrium hot electrons accomplish the physical picture.

We have realized a plasmonic interferometer formed by a nanoslit and a nanogroove in a single-crystal gold film. The possibility of measuring laser pulses of ultimately short durations, corresponding to two periods of a light wave (6 fs pulse duration), has been demonstrated using this interferometer.

At present, the measurement of ultrafast, on a femtosecond time scale, optical fields with a nanometer spatial resolution still seems to be a challenging problem. These measurements are fundamentally important both for the comprehension of a spatio-temporal dynamics of ultrafast pulses [1] and for many scientific and practical applications, such as an investigation of the interaction of light and matter on a femto/nano scale [2]. Measurement of ultrafast optical pulse with a nanoscale spatial resolution is even a more challenged problem. Only several methods are known. Both are rather difficult to realize experimentally and involve a complicated mathematical processing, which impose considerable limits on their application in practice.

Figure 1. Schematic diagram of the setup for the measurement of spatio-temporal properties of femtosecond laser radiation using a single plasmon SHR nanostructure.
We propose and describe a new concept of a nanoprobe for the characterization of femtosecond laser pulses, which is based on a giant optical nonlinearity of plasmonic nanostructure [3] (Fig. 1). In the upshot we demonstrate a nanoscale size, ultrafast, and multiorder optical autocorrelator with a single plasmonic nanostructure for measuring the spatio-temporal dynamics of femtosecond laser light. As a nanostructure, we use a split hole resonator (SHR), which was made in an aluminum nanofilm. The Al material yields the fastest response time (100 as). The SHR nanostructure ensures a high nonlinear optical efficiency of the interaction with laser radiation, which leads to (1) the second, (2) the third harmonics generation and (3) the multiphoton luminescence, which, in turn, are used to perform multi-order autocorrelation measurements [4]. The nano-sized SHR make it possible to conduct autocorrelation measurements (i) with a subwavelength spatial resolution and (ii) with no significant influence on the duration of the laser pulse. The time response realized by the SHR nanostructure is on the time scale of about 10 fs.

We demonstrate femtosecond plasmonic interferometry with a geometry slit – groove on the monocrystalline Au(111) surface. The plasmonic microinterferometer consists of a tilted slit-groove pair. This arrangement allows us to perform: (1) the measurements of real and imaginary parts of the metal dielectric function of Au(111) nanofilm, and (3) the measurements of the femtosecond laser pulse duration in several periods of light oscillations (6 fs).

References:


SURFACE MICROCAVITIES AT NANOSCALE DEPTHS PRODUCED BY ULTRAFAST LASER PULSES

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The evolution of silicon surface (111) induced by a single femtosecond laser pulse (350 fs, 1028 nm) at near-threshold fluences in viscous liquid is investigated by SEM and AFM. Depending on the deposited energy, the near-surface layers undergo several transformations that generally resulted in controlled formation of simple and complex bowl-shaped surface microcavities at nanoscale depths, surrounded by characteristic rims. At fluences below the ablation threshold, a 2-nm-depth microcavity is observed. The embedded microcavity has the form of an ultra-smooth paraboloid with a roughness ~0.1 nm rms. Its formation is associated with a flow of melt from the center to the edges due to surface tension variations induced by a single Gaussian shaped femtosecond laser pulse above the melting threshold. Similar microcavities with depths up to 4 nm are also observed with an increasing fluence. However, at higher fluences complex microcavities, characterized by a central sub-nanostructured region (with a roughness ~0.5 nm rms) encircled with a crown-like rim of ~5 nm height and ~50 nm width (at FWHM), are found. The carried out experiments clearly demonstrate the surrounding medium to play a crucial role in producing of smooth and nanostructured surface microcavities at nanoscale depths with single femtosecond laser pulses.

UV NONLINEAR OPTICS WITH SINGLE PLASMONIC NANOSTRUCTURE

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Abstract: We report a strong nonlinear optical interaction of a single plasmonic nanostructure - Split–Hole Resonator (SHR) - with a two light periods laser pulse that leads to (i) the second harmonic generation, (ii) the third harmonic generation (THG), and (iii) the light generation at mixed frequencies. The THG near field amplitude reaches 0.6% of the fundamental frequency field amplitude, which enables the creation of UV radiation sources with a record high intensity. The UV THG may find many important applications including biomedical ones (such as cancer therapy).
1. Motivation

Due to the extremely weak optical response of a single nanostructure, until recently, the main research in nanoplasmonics was carried out with ensembles of nanoparticles (typically 10^4-10^5 nanostructures) [1]. In the ensembles of nanoparticles both structural and material parameters usually vary from one nanoparticle to another, so the optical response is averaged over the ensemble. On the other hand the current state of experimental optical and spectroscopic technique allows to carry out measurements at the level of single atoms and molecules [2]. Measurements on the level of individual nanostructures is an effective method for studying the fundamental optical and spectroscopic properties of nanostructures, allowing one to define the mechanisms of elementary physical processes and to avoid the inevitable averaging over the ensemble with the inevitable loss of physical information about the parameters of nanoparticles, making development of optical methods for the detection and characterization of individual nanostructures to form a central place in the basic and applied nanotechnology research.

![Fig. 1. Schematic representation of the use of nano-localized ultraviolet light source for the treatment of a living cell by a single Au nanostructure [3]](image)

2. Results

This work presents our results of investigation of nonlinear optical interaction of a single plasmonic nanostucture - Split–Hole Resonator- with a two light periods laser pulse to realize maximum conversion efficiency of third harmonics generation in the UV spectral range [3]. The strongest THG emission was realized with a single gold nanostructure in a geometry of the Split-Hole Resonator under the state-of-the-art experimentally realized conditions. All spectral components of a 6 fs laser pulse excite simultaneously several multipole plasmon resonances in the SHR nanostructure. We report a strong nonlinear optical interaction at the frequencies of these resonances that leads to (i) the second harmonic generation, (ii) the third harmonic generation (THG), and (iii) the light generation at mixed frequencies. The THG near field amplitude reaches
0.6% of the fundamental frequency field amplitude, which enables the creation of UV radiation sources with a record high intensity. The UV THG may find many important applications including biomedical ones (such as cancer therapy).

3. References


FABRICATION AND OPTICAL CHARACTERIZATION OF EPSILON-NEAR-ZERO VIS-METASURFACES

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By the metod of magnetron sputering we made two-layer thin translucent composite films. Deposition of films was carried out on fresh samples of commercial silica glass in Mini Sputter Coater Quorum Technologies SC7620. Deposition occurred in an argon atmosphere (pressure ≈ 20 Pa, residual air pressure ≈ 2 Pa) by magnetron sputtering of Cu and Ag foils. The first layer is Ag film with the thickness 5-10 nm deposited for 15 seconds. Unlike silver, sputtering a copper target in the presence of residual air pressure led to the deposition of dicopper oxide. So the second layer is dicopper oxide film with the thickness 5-10 nm. The time of deposition was changed from 30 to 45 seconds to change the ratio of the metallic and oxide components.

Composite film has an islet structure (fig.1a), translucent, in transmission has a golden color and has a metallic shine (fig.1b).

Fig. 1 SEM (a) and optical (b) Image of composite film
This reflection and transmission spectrum of composite, silver and copper oxide films (fig.2) shows near zero reflection of the composite films (at the level of the glass substrate).

The position of the reflectance minimum of the composite is expected to shift to the "red" region from (four hundred and sixty) 460 nm (c1) to (four hundred and ninety) 490 nm (c2) - as the proportion of the dicopper oxide in the composites increases. Near-zero reflection of composite films can be explained by near-zero value of the reflection coefficient.

Fig.2. The spectrum of the reflectance (R) and transmittance (T) coefficients for the glass substrate (s), for Ag film (m, metal), copper (I) oxide film (d, dicopper oxide) and composite films (c1, copper oxide deposition time - 30 s) and (c2, copper oxide deposition time - 45 s).

LARGE-SCALE FABRICATION OF SULFUR-HYPERDOPED SI NANOSHEET ARRAYS VIA ULTRAFAST LASER SURFACE NANOTEXTURING

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Large-scale surface nano- and micro-texturing of commercial Si wafer in the form of nanometer-thick vertical sheets inside micro-craters was performed during multi-shot IR picosecond laser ablation under a 2-mm thick carbon disulfide liquid layer. The textured surface
layer demonstrates broad ultralow mid-IR transmittance and considerable content of sulfur, carbon and oxygen in the 200-nm thick layer of surface ripples. High-resolution TEM studies indicate the anti-correlating abundance of the impurity elements and the different crystalline structure of ripple trenches and edges, enlightening the underlying ripple formation and sulfur doping mechanisms.

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References

Chair L.V. Seleznev

DYNAMICS AND SPECTRA OF LIGHT BULLETS IN TRANSPARENT DIELECTRICS.

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A light bullet (LB) is a short-lived formation in a femtosecond filament with a high spatiotemporal light field localization. The formation of LBs in the result of simultaneous laser beam self-focusing and pulse selfcompression in transparent dielectrics is a bright phenomenon in femtosecond filamentation. A necessary condition for implementing this regime is the existence of anomalous group-velocity dispersion. We present the results of experimental and numerical investigation of LB dynamics in femtosecond filament. The scenario of the LB formation in the presence of anomalous group velocity dispersion is presented within the same general scenario for condensed matter and humid air. The temporal and spectral parameters of LBs during filamentation in fused silica, different fluorides crystals and humid air are obtained. The sequence formation of the quasiperiodical LB is obtained numerically and is confirmed experimentally by autocorrelation measurements of the LB’s duration. The estimation of the LB duration reaches few-cycle value, its lifetime is about several picoseconds and the peak intensity achieves value of $10^{14}$ W/cm$^2$. The difference between group and phase velocity of wave packet in dispersive medium lead to a periodic change of light wave’s absolute phase, which causes oscillations of light field’s amplitude and the radius of LB during the propagation in the filament.

It is established that the generation of each LB is accompanied by the ejection of a supercontinuum (SC) in the visible spectrum and an isolated anti-Stokes wing is formed in the visible area of the SC as a result of destructive interference of broadband spectral components. It was found that the energy of a visible SC increases discretely according to the number of LBs in the filament. A giant blue shift (more than 3000 nm) of an isolated visible band of supercontinuum was discovered and studied in the single filament regime of Mid-IR femtosecond laser pulse at powers slightly exceeding critical power for self-focusing in fluorides. We demonstrated that the model of ionization in solid dielectric which is used in numerical simulation fundamentally affects the obtained scenario of LB formation. In the result of experimental and analytical investigation it is revealed that material dispersion and multiphoton order determine main peculiarities of anti-Stokes supercontinuum band shift under filamentation of near- and mid-IR fs pulses at strong anomalous GVD in transparent dielectrics.
A law determining the dispersion shift of the anti-Stokes band of the supercontinuum of a light bullet in a filament of a femtosecond laser pulse in transparent dielectrics has been established. The dispersion equation theoretically obtained for the anti-Stokes shift has been confirmed by spectroscopic studies of the filamentation of the near and mid-IR ranges in fused silica and fluorides.

**FORMATION OF A HIGHLY DIRECTIONAL SUPERCONTINUUM UPON FILAMENTATION OF RADIATION IN AIR**

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Presently, studies of a supercontinuum (SC) in a visible spectrum range (white light laser) upon filamentation of the laser pulses with femtosecond duration in air are a great interest. It is due the availability and great practical significance of this type of radiation for solving tasks in metrology, telecommunications, nanotechnology, optical coherence tomography, remote analysis of the atmosphere, and many others. In this paper we research the conditions of the laser beam filamentation in air and a highly directional SC formation in the visible spectrum range.

A solid-state starter complex "Start 480" was used in the experiments. The complex includes a Ti:Sa master oscillator with a continuous pump laser, optical stretcher, one regenerative and two multi-pass amplifiers with pulsed pump lasers and compressor on two diffraction gratings. The output radiation parameters were as follows: a central wavelength of 940 nm, spectral width of 26 ± 2 nm (FWHM), pulse duration of 70 ± 3 fs, energy up to 50 mJ, beam diameter of 10 mm. The complex operated with a pulse repetition rate of 10 Hz, and the energy stability was 3%. The output radiation was linearly polarized (horizontally), and the beam quality factor M2 = 2.

Focusing of radiation carried out by a spherical mirror at the different focal length, incidence angles of radiation on the focusing mirror and with a different numerical aperture of focusing system (NA). It is shown that at a low numerical aperture (NA ≤ 1.5×10^-3) Kerr nonlinearity plays a decisive role in the formation, existence and termination of the filament. The reasons for the influence of the Kerr nonlinearity are discussed in the report. It is shown that the SC radiation is most stable when formed in the presence of aberrations in the wave front of the laser beam. Optimal aberrations realized at the incidence angle of 150 on focusing spherical mirror with F = 744 mm. Appearance this radiation occurs after visible filament through a gradual conversion of the spectral composition from long wavelength to short wavelength (to 350 nm). On the track section 35–135 cm from the filament, radiation propagates in the form of a spatially stable structure similar...
to a soliton with a transverse dimension \( \leq 300 \ \mu m \). In this case, SC radiation is significantly different from conical off-axis emission occurring in the aberration-free filament: it has a divergence close to the diffraction limit, linear polarization, and a shorter-range wing of the spectrum. The infrared component of SC has the radiation duration of 25 fs. Research results are presented and forming mechanism of such radiation is discussed.

DIFFRACTION-RAY OPTICS OF “DRESSED” LASER BEAM FILAMENTATION IN AIR

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Self-action in air of high-power femtosecond laser pulse in the spatial form of a ring-Gaussian beam ("dressed" beam) is studied theoretically. Pulse self-focusing and filamentation is analyzed in the detail through the numerical solution of the spectral propagation equation taking into account medium optical nonlinearity and plasma generation. Pulse propagation dynamics and energy flows inside the beam are visualized by means of the averaged diffraction rays tracing. We clearly show that in terms of diffraction optics, the outer ring forms a diffractive waveguide favoring long-range self-channeling of the central part of a beam delivering optical energy to a filament. The spatial robustness and stability of this diffractive waveguide strongly depends on the energy stored in the ring, as well as on its position relative to the beam axis. The striking advantage of the “dressed” beam is also its reduced angular divergence during plasma-free (post-filamentation) evolution.
We investigate the guiding of electrical discharge using femtosecond laser filamentation in air. A feature peculiar of the guided discharge is its straight line geometry (Fig.1).

Generally, the duration of a guided discharge stimulated by femtosecond laser filamentation is defined by the high voltage source. In our guided discharge experiments a Tesla Coil generator was used to generate voltage pulse of 360-kV. The discharge duration is defined by the product of the Tesla geometrical output capacitance and the load resistance. In our case both the circuit resistance and the discharge resistance contribute to the load resistance. As a result the discharge duration is shorter than 1 mks in our case [1].

For several applications such as the virtual plasma antenna or the capture of high current, a guided discharge of much longer duration is required.

We present here experiments carried out with a discharge length of 100-cm. To increase the duration of the laser guided discharge we employed a second circuit which injects an additional current pulse under much smaller voltage. Our first experiments with prolongation of 85-mm discharge up to 130 mks were presented in [2]. With help of high speed imagery we showed that 100-cm discharge guidance has been achieved successfully during more than 1 ms.

Fig.1 A guided discharge image


Remote detection of oil spills on the water surface is a very important issue for environmental monitoring. Nowadays there are several facilities using laser spectroscopy [1, 2] for this purpose. They operate in near-UV or visible wavelengths domains [1, 3]. In this work, we propose excitation of the fluorescence with ultrashort UV pulses. Due to self-focusing and following filamentation [4] diffraction is suppressed and high intense emission can be delivered to a distant target. We conducted the model experiment on detecting the fluorescence of oil products thin films on water surface excited by femtosecond UV laser pulses.

Pulses of second (372 nm) and third (248 nm) harmonics of femtosecond (100 fs) Ti:Sapphire laser facility were focused with 20 cm lens to the thin films of oil products (crude oil, oil VM-5, oil 5W-40 and solvent WhiteSpirit) on the water surface. Luminescence was collected to a spectrometer slit via BaF\(_2\) lenses. Luminescence spectra of all considered pollutants had unique and well distinct shapes and were extended from 300 to 400-700 nm, so they were isolated from the laser wavelength for 248 nm excitation and overlapped with scattered laser emission for 370 nm case. Moreover, oil VM-5 and WhiteSpirit were transparent for 372 nm pulses. Thus, pulses of 248 nm wavelength were shown to be preferable for fluorescence excitation.

High level of luminescence signal is crucial for long range detection of oil spills. We observed increase of overall fluorescence signal with pulse power and following saturation at the critical power for self-focusing. To increase luminescence signal multiple filamentation may be used, because in this case every filament independently contributes to the total fluorescence signal.

Thus, we experimentally studied applicability of UV femtosecond laser pulses for remote detection of oil and oil products films on water surface. Shorter UV wavelengths were shown to be more suitable for fluorescence excitation. Multiple filamentation can lead to increase of luminescence signal. This work was supported by RFBR grants 14-02-00489 and 14-22-02021, LPI Educational-Scientific Complex, RF President grant MK-5795.2015.2


[2]. V. V. Fadeev, S. A. Dolenko, T. A. Dolenko, Ya. V. Uvenkov, E. M. Filippova, V. V.


PERCULARITIES OF REGULIRIZED FILAMENTS EVOLUTION AND SUPERFILAMENTATION IN AIR


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In the talk we discuss the results of our experiments on multiple and super filaments control by means of modifications of femtosecond laser pulse wave front. The considering regime of filamentation is reached by weakly focusing (f=3m) a terawatt femtosecond laser pulse (55 fs, 30mJ, 800 nm) in air and introducing amplitude and phase masks transversely to the initial laser beam. The diagnostic is based on the single-shot measurements of frequency-angular spectrum of radiation and registration of acoustic signal by broadband piezoelectric transducer at different positions along the filament. In the focal region, a reorganization of the interacting filament bundle takes place with the emergence of a new filamentary structures with a higher intensity, wider spectrum etc.

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High power ultrashort UV pulses have a broad range of applications – from high-voltage electric discharge initiation to environmental monitoring. UV pulses of high peak power are usually obtained amplifying a seed pulse in gas laser amplifiers. Passing through bulk optical elements the pulse undergoes temporal elongation and phase modulation. Number of optical elements differs with required amplification, therefore resultant pulse parameters varies. However, knowing pulse parameters is essential for conducting experiments, correct interpretation of the results and comparison with numerical simulations. In this work, we discuss methods of measurement of amplified pulse duration, critical power of self-focusing and power density transverse distribution. We applied these measurement techniques to studying of regularized sub-TW sub-ps UV laser pulse propagation in air in filamentation regime. Sub-TW sub-ps laser pulse was obtained by amplification third harmonics (248 nm) of Ti:Sa laser system in e-beam excimer KrF amplifiers [1]. Pulse duration was measured with single-shot XeF autocorrelator [2] and amounted to approximately 900 fs. We obtained power density transverse distribution of wide-aperture UV beam collecting glass screen luminescence during interaction with UV beam to CCD-camera by an objective lens. The luminescence of the glass is nonlinear in dependence on pulse power, so we calibrated it to reconstruct the energy density distribution and increase the contrast of CCD-images.

Critical power of self-focusing for studied laser emission is crucial for filamentation investigation. Concerning amplifies laser pulses, critical power of self-focusing can decrease for stretched and modulated in pass-through optics pulses. We measured it directly for different pulse durations. Plasma distribution along the channel generated by focused laser pulse depends on pulse energy. Pulse with energy, higher the one, corresponding the critical power of self-focusing, creates asymmetrical plasma channel, unlike pulse with lower energies. So, we measured critical power of self-focusing for pulses of different durations.

We use measured above parameters to characterize of the beam in the experiment on filamentation regularization [3]. We observed 15 meters long continuous filaments in case of
regularization. Calibration of energy density distributions allowed us to calculate filament radius. Also, we estimated energy in one hole of the mask and compared it with critical power.

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SPATIOTEMPORAL DYNAMICS OF OPTICAL VORTEX IN FEMTOSECOND MID-IR FILAMENT

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Femtosecond filamentation has been extensively studied for beams of different amplitude and phase distributions [1, 2], including Airy and Bessel beams [3]. Filamentation with optical vortex is studied significantly less. Meanwhile zero laser field of vortex at optical axis tends to formation of spatial tubular structures in filamentation [4], which can be used to create modifications of cylindrical shape in transparent solid dielectrics. We numerically investigate a spatiotemporal dynamics of mid-IR filamentation of circular beam with phase singularity in CaF$_2$. A characteristic feature of this wavelength range is negative dispersion of the pulse group velocity, which can result in formation of «light bullets» [5].

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

$$2i k_0 \frac{\partial A}{\partial z} = \hat{T}^{-1} \left[ \left( \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) - \frac{m^2}{r^2} \right) A \right] + \hat{D}[A] + \frac{2k_0^2}{n_0} (\Delta n_k - \Delta n_{pl}) A - ik_0 (\sigma N_e + \alpha + \delta) A,$$

$$\frac{\partial N_e}{\partial \tau} = R_E (N_0 - N_e) + N_e (\nu_i - \beta).$$

The propagation equation includes operator of wave non-stationarity $\hat{T}^{-1}$.

We studied filamentation of the optical vortex in bandwidth-limited pulsed doughnut laser beam with gaussian envelope:
\[ A(r, t, 0) = A_0 \left( \frac{r}{r_0} \right)^2 e^{-r^2/2r_0^2} e^{-t^2/2t_0^2} e^{i m \varphi} \]

Parameters of the pulse were chosen as \( m = 2, r_0 = 230 \, \mu\text{m}, t_0 = 60 \, \text{fs}, \lambda_0 = 3000 \, \text{nm}, P = 6P_{cr} \), where \( P_{cr} \) is a critical power for optical vortex [6].

At the initial stage of propagation the laser pulse is self-focusing in a thin ring at the distance 1.7 cm (Fig. 1). Dependence of the linear plasma concentration has a notable peak at this distance \( (\rho \sim 4 \times 10^{13} \, \text{cm}^{-1}) \), fluence becomes higher than 0.12 J/cm². Self-focusing continues until plasma nonlinearity starts to defocus obtained tubular structure. Simultaneously diffraction makes energy to flow towards optical axis. In combination with further refocusing this results in formation of two rings in pulse cross-section at distance \( \sim 3 \) cm.

![Fig. 1. Fluence distribution \( F(r, z) \) and linear plasma concentration dependence on propagation distance \( \rho(z) \)](image)

These rings move towards each other up to at least 4.5 cm, shifting in time to trailing part of the pulse due to relatively small group velocity. Note that the plasma concentration \( \rho(z) \) reaches its maximum in the vicinity of the first ring focus, after which it will not reach noticeable values.


THE EFFECT OF WAVEFRONT CURVATURE ON SUPERCONTINUUM GENERATION UNDER NEAR-IR FEMTOSECOND FILAMENTATION IN WATER

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We experimentally and numerically studied two regimes of femtosecond filamentation in water, differed by the water sample position relative to the focal plane of the focusing lens: in the first case water sample is placed after the focal plane (case (1)) and in the second water sample is placed before the focal plane (case (2)). We demonstrate that using spatially divergent incident femtosecond 1240-nm laser pulses in water (case (1)) leads to an efficient supercontinuum generation. We found that the optimal conditions for supercontinuum generation are realized when the focal plane is placed 100 – 400 μm before the water surface. In comparison with case (2) the gain in supercontinuum energy could achieve 100 (NA=0.5 E=10μJ). The effect is stronger for tight focusing conditions and low laser energies. If the focusing is sufficiently weak (NA<0.2), the lower the laser pulse energy is (the closer it is to the filamentation threshold), the higher the SC energy generated in divergent beams is in comparison to the SC energy generated in convergent beams. Under strong focusing conditions (NA=0.2) and high-energy laser pulses, the supercontinuum generation is suppressed for convergent beams in contrast to divergent beams that nevertheless are shown experimentally to allow supercontinuum generation.

The analysis by means of three dimensional unidirectional pulse propagation under sufficiently weak focusing conditions shows that compared to filamentation of laser pulses in convergent laser beams, the laser-induced plasma is less dense leading to a longer filament and thus to more efficient energy transformation into supercontinuum radiation. Thus, the focusing geometry of the incident pulse in the spatially divergent incident femtosecond laser pulses supports an effective nonlinear conversion of the main frequency to the SC spectral components propagating along the axis, which is highly demanded in femtosecond optical parametric amplifiers.
Light bullet is a wave packet, extremely compressed in time and space in the nonlinear optical interaction of femtosecond radiation in the anomalous group velocity dispersion. Light bullets with high spatial and temporal localization of optical radiation is a new object of research in spectroscopy and laser physics. The light bullet peak intensity reaches $10^{13} - 10^{14}$ W/cm$^2$, its duration is 1-2 optical cycles and diameter reaches several wavelengths.

A study of the general laws of formation and dynamics of wave packets of several optical oscillation cycles duration is a little-known fundamental problem of modern nonlinear optics, the solution of which is significant for a wide range of problems of interaction of electromagnetic radiation with medium. Nonlinear interaction of such extremely localized wave packet with the media essentially depends on phase relations of the light field and its envelope, which are extremely important for the modern attosecond optics. For these packets one can also realize direct registration of the phase of the light field within the packets. This opens up novel possibilities for fundamental studies of how the phase relationships affect the nonlinear optical interaction of these packets with the media.

Novel in experimental studies of the compressed wave packets dynamics is the laser coloration method in the lithium fluoride (LiF) crystal. Opposite to on-line experiments where the registration of supercontinuum radiation and plasma emission is inevitable, the structure of extremely compressed wave packet is recorded via appearance of the color centers in LiF, and may be investigated much later by measuring the luminescence of color centers in the weak pump field. The color centers are created on a picosecond timescale, so they do not influence the femtosecond pulse dynamics. Laser coloration method allows to investigate in details the 3D structure of the light bullet optical field with a spatial resolution better than one micron (determined solely by the resolution of the microscope used) after just single pulse passed through the material. This feature, as well as the extreme ease of laser coloration method, distinguishes it from the most advanced methods of light bullet dynamics study, such as 3D imaging technique. Such an experiment allowed us to unambiguously answer the debated question about light bullet path length.

Also we propose the laser coloration method to investigate the waveguide regime of laser pulses self-organization. This regime is developed due to the increase of the LiF refractive index on the
beam axis after the train of pulses passes through and create color centers. The induced color centers structure increases with exposition: single-pulse create a 1-mm-long wavelength-wide string of color centers, but after several thousand pulses this waveguide can increase its size by a factor of ten and more. The presence of the waveguide produced by induced color centers can significantly affect the process of light bullet self-organization, increase light bullet propagation length, change the frequency-angular spectrum of radiation qualitatively, and shift anti-Stokes wing of supercontinuum to shorter wavelength.

SIMILARITY CRITERION FOR LIGHT BULLETS AND SUPERCONTINUUM FORMATION UNDER FEMTOSECOND FILAMENTATION IN AGVD CONDITIONS.

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Using experiment and numerical simulation, we study the effect of anomalous group velocity dispersion (AGVD) on the formation of a visible supercontinuum (SC) band and light bullets (LB) arising in the course of filamentation of mid-IR femtosecond pulses in fused silica and fluorides. It is found that the anti-Stokes shift of the visible band increases with increasing energy of the pulse, the centre wavelength of which lies in the region of weak AGVD, and does not depend on this energy in the region of strong AGVD. A criterion is introduced for assessment of the AGVD ‘strength’, at which the stable visible supercontinuum band accompanies the formation of a robust light bullet in the mid-IR filament.

Basing on the analysis of phase relations for harmonics of broadband SC emitted by the LB, it is found that the anti-Stokes shift of isolated wing of SC is determined by the material dispersion of the medium. The dispersion equation for the anti-Stokes band of LB’s spectrum is obtained. It allows to calculate analytically the magnitude of the shift depending on the pulse’s wavelength and the material dispersion of the medium. The rule established theoretically has an experimental confirmation by the spectroscopic studies of the SC during filamentation in fused silica and fluorides of femtosecond pulses at the wavelength, varying in near- and mid-IR ranges. Thus, for any dielectric material with known dispersion dependence the position of the anti-Stokes’ wing maximum in spectrum of supercontinuum could be predicted.
THE INFLUENCE OF WAVE PACKET’S SCALES ON FEMTOSECOND FILAMENTATION AND FORMATION OF LIGHT BULLETS

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During filamentation of femtosecond wave packets in transparent dielectrics under conditions of anomalous group velocity dispersion (AGVD) occurs a robust formation with high spatio-temporal localization of light field – light bullet (LB). The formation of light bullets takes place under simultaneous and symmetrical compression of wave packet in space and time [1]. We have investigated the influence of ratio between characteristic scales (dispersive ($L_{\text{disp}}$) and diffraction ($L_{\text{dif}}$) lengths) of wave packet on femtosecond filamentation and LB formation in condensed matter.

We consider femtosecond laser wave packet at the wavelength $\lambda_0=2000$ nm with $\tau_0=30$ fs ($L_{\text{disp}}=9$ mm) during filamentation in fused silica. Varying the radius of a beam $L_{\text{dif}}$ has been changed more than in 100 times in conditions of constant $L_{\text{disp}}$. The ratio of diffraction and dispersion lengths varies in wide range ($10^{-3}$–$10$), that allows to investigate the degree of AGVD’s influence on femtosecond filamentation and LB formation. Under variation of pulse energy, numerically was detected threshold power of filamentation for different characteristic length ratio.

If $L_{\text{disp}} \ll L_{\text{dif}}$, then the decrease of intensity during dispersive spreading of a pulse on an initial stage of propagation leads to an increase of threshold power of filamentation. Moreover, the distance before the start of a filament is greater than the stationary self-focusing length.

When $L_{\text{disp}}$ and $L_{\text{dif}}$ are close, pulse self-compression and beam self-focusing are coordinated. Joint pulse compression in space and time leads to decrease in threshold power of filamentation in comparison with critical power of stationary self-focusing. Furthermore, the distance before filament emergence decreases compared to length of stationary self-focusing.

If $L_{\text{disp}} \gg L_{\text{dif}}$, then the threshold power of filamentation and the distance before filament start could be detected as in stationary case.

Independently from the ratio between $L_{\text{dif}}$ and $L_{\text{disp}}$, the peak intensity in LB, achieves 60–65 TW/cm² and this fact confirms its robustness.
Plasma density spatial distribution and plasma decay in a filament produced by an intense femtosecond laser pulse were studied in air and other gases (nitrogen and argon) for different pressures and external electric fields. Measurements by means of transverse optical interferometry showed that in air at atmospheric pressure the maximum plasma density in the filament reached the value of \(~10^{17} \text{ cm}^{-3}\) with filament diameter of about 150 \(\mu\text{m}\). Decreasing the pressure down to 20-50 Torr was not accompanied by a decrease in plasma density, while the filament diameter reduced by 20-30\%. At lower pressures plasma density started to decrease, while the diameter did not change significantly. Similar results were obtained for N\(_2\) and Ar (but with filament diameter of \(~80 \mu\text{m}\) at atmospheric pressure). It was demonstrated by means of the terahertz scattering technique that plasma density in air and nitrogen at atmospheric pressure was reduced by two orders of magnitude within 3–4 ns with the decay rate decreasing with decreasing pressure. The argon plasma density remained stable within several nanoseconds for the 50–760 Torr pressure range. Application of external electric field resulted in slowing down of plasma decay: for the electric field of 7 kV/cm the lifetime of plasma with the density above 10\(^{16}\) cm\(^{-3}\) was increased from 0.5 ns to 1 ns. Numerical simulation of electron density dynamics and electron temperature evolution was performed taking into account dissociative and three-body electron-ion recombination, formation of complex positive ions as well as evolution of electron temperature. The simulation showed that when an external electric field was applied, the electron temperature evolved nonmonotonically and passed through a minimum due to electron heating during the electron-ion collisions. The initiation of a pulsed microwave discharge in atmospheric air by a plasma filament induced by intense femtosecond laser pulses was also studied. It was shown that the electric field threshold for the initiated discharge was reduced by about a factor of two, from 25 to 12 kV/cm, compared with the self-discharge. Channelling of the atmospheric-pressure microwave discharge along the plasma filament was observed. The lifetime of the initiated discharge plasma was determined by the duration of the microwave pulse (\(\sim 1\–2\ \mu\text{s}\)) with the...
maximum electron density estimated to be $4 \times 10^{15}$ cm$^{-3}$ (for microwave frequency $f = 36$ GHz). The developed theory of the microwave radiation propagation along the plasma channel created by a femtosecond laser pulse predicted that the relatively low conductivity of the laser plasma and its rapid decay caused the limitation of the initiated microwave discharge propagation length to a few centimeters.

**MULTIPLE FILAMENTATION OF SUBPICOSECOND TERAWATT UV LASER BEAM AND ITS POSSIBLE APPLICATIONS**

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Transparent medium polarization in intensive laser light causes global radiation self-focusing or a small-scale beam fragmentation into a number of quasi-stable filaments if laser power exceeds the critical value $P_{cr}$, determined by a balance with beam diffraction. TW-level peak power in subpicosecond UV pulses produced at the Ti:Sapphire – KrF laser facility GARPUN-MTW is 4 orders of magnitude higher than $P_{cr} \sim 0.1$ GW [1]. Such supercritical laser beam is unstable: “hot spots” in intensity distribution provokes laser beam break up into multiple filament-like light channels, being managed by a coherent rotational stimulated Raman scattering at atmospheric nitrogen molecules [2]. Increased intensity and energy fluence in filaments is responsible for shortpulse energy saturation in KrF gain medium and nonlinear losses in laser windows [3]. On the other hand, there is a big room for practical applications of the UV beam filamentation, e.g. for triggering and guiding HV discharges, directed transfer of MW radiation in virtual plasma waveguides, and excitation of lasing in plasma filaments. In the present communication, we are reporting the following achievements in multiple filamentation study at GARPUN-MTW facility:

- Effective suppression of multiple filamentation of the beam in Xe by exploring a 2-photon resonance of KrF laser radiation with $6 \text{p} [1/2]_0$ state, which resulted in filament de-focusing due to a large negative nonlinear refractive index. It was accompanied by monochromatic narrow-angle coherent cone emission at 828-nm wavelength [4].

- Arrangement of a few hundred randomly distributed filaments into well-organized 2D filament array by using periodic amplitude masks with circular or square apertures of 5-mm size. Being set 5-m beyond the final amplifier masks introduced controllable periodic perturbations in the beam via linear Fresnel diffraction that overcame occasional perturbations [5].
Atmospheric air lasing at 1.07-μm wavelength with radiation divergence ~ 1 mrad and a small-signal gain ~ 0.5 cm⁻¹ at molecular nitrogen transition in a super-filament carrying intensity ~ 5·10¹⁴ W/cm² produced by focusing of a bundle of UV filaments.

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LECTRON-FREE FILAMENTATION OF THE ULTRASHORT LASER PULSE DRIVEN BY COHERENT STIMULATED RAMAN SELF-SCATTERING

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In spite of many decades of intensive studies in the physics of filamentation, a number of specific features of this phenomenon are still under debates. One of these features is the so-called non-ionizing filamentation effect which for the first glance is in contradiction with conventional model of filamentation considering balance of Kerr self-focusing and plasma defocusing. Non-ionizing (or electron-free) filamentation has been reliably established in a number of experiments [1,2]. Attempts to explain the nature of this effect are now related to the higher order terms of the Kerr nonlinear index of refraction of air in a series expansion with alternate positive and negative signs [1,2].

Role of the stimulated Raman scattering (SRS) on the rotational states of molecules in the filamentation of ultrashort laser pulses in air has been well studied [2]. However, it turns that to fit theory with experimental results one should assume the value of the characteristic transverse relaxation time T2 being of the order of ~100 fs [3]. One should stress that this value is in direct contradiction with conventional physics of atomic and molecular collisions which predicts three order larger value, of ~100 ps.

In this report we propose the coherent stimulated rotational Raman self-scattering as a mechanism of electron-free filamentation of the ultrashort laser pulse in air. Coherent SRS arises when the pulse width τp is shorter than the characteristic relaxation time T2 and the pulse spectrum
is wider than the eigen frequency $\Omega$ of the equivalent two-level model oscillator (i.e., the Stokes shift) [4]. We analyze main features of coherent SRRS-driven filamentation in air using the envelope equation approach [5]. Evolution of laser pulse is then guided by the normalized fluence. Nonlinear oscillations in the equivalent two-level system result in nonlinear polarization which determines the pulse spectrum transformation and the self-focusing effect. When self-focusing starts, the normalized fluence increases and reaches the value $W \approx \pi/2$ and nonlinear polarization changes its sign which results in defocusing. Thus, filament radius exhibit oscillations near this equilibrium state, and the filament is formed without plasma refraction. The theory developed provides us to explain our recent experimental results on electron-free multiple filamentation produced by propagating along 100-m distance 1-ps sub-TW pulses of UV ($\lambda=248$ nm) radiation from GARPUN-MTW Ti: Sapphire / KrF laser facility [6,7].

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Filamentation of laser radiation is the key effect of interest in the problems of femtosecond atmospheric optics because it is associated with such phenomena as the localization of light energy, the formation of extended plasma channels, generated of supercontinuum, etc. Study of filamentation in environments with strong cubic nonlinearity are interest, both in terms of modeling of terawatt laser pulses on long atmospheric paths and to solve problems in micro-optics, for modifying the properties of materials. Experiments on the filamentation of laser beams in air are shown that the change of the beam diameter and its initial focusing allow to effectively control the position of the multiple filamentation zone on the scale of hundreds of meters. The defocusing of the beam, as an instrument for removing the filamentation zone from the source, has limit levels that depend on the radius of the beam and its power, exceeding which leads to the termination of filamentation. Controllable filamentation of the laser beam makes it possible to form at a given distance from the source the values of the optical field intensities sufficient to induce plasma on the targets for analyzing their elemental composition. In experiments on filamentation of laser beams with a Gaussian profile in a medium with strong Kerr nonlinearity, it is shown that with increasing pulse power the region of multiple filamentation increases in length and diameter, the distribution of filaments in the longitudinal direction of the filamentation zone has a maximum. When the power reaches > 105 Pcr the filamentation zone acquires the shape of a hollow cone directed toward the source of the laser radiation. The average length of individual filaments is reduced when the pulse power is increased while maintaining an almost identical diameter. In experiments on multiple filamentation of laser beams in media with different Kerr nonlinearity (air, water, glass) revealed a quantitative relationship between the initial laser beam parameters (energy, power, diameter, focus) with the characteristics of the of multiple filamentation zone (number of filaments, their distribution in longitudinal and transverse direction, the length of multiple filamentation zone, its position on the track, the length of individual filaments, the transformation of the spectrum).
SPATIAL AND SPECTRAL CHARACTERISTICS OF THE POST-FILAMENTATION ZONE FEMTOSECOND LASER PROPAGATION PULSES ALONG THE PATH

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The propagation of laser radiation of ultrashort duration upon reaching critical values of the pulse power \( P_{cr} \) is accompanied by its self-focusing and filamentation. In this case, significant changes occur in the energy, spatial, spectral, and angular characteristics of laser radiation. The radiation energy is localized in narrow light filaments containing a pre-threshold density of light energy flux, the spectral composition of the radiation is substantially enriched. These properties have priority values for atmospheric optics problems - laser sounding of the atmosphere and transportation of localized light energy through the atmosphere. At the same time, the task of translating the filamentation zone to a considerable distance from the source is necessary, and for the atmospheric optics, the most attractive are the multiple filamentation of the beam.

The processes of formation of the transverse structure of a laser beam after multiple filamentation were investigated. It is shown that the spectra of post-filamentary light channels, rings and beam differ substantially. The spectrum of post-filamentation channels has a significant and symmetrical spectral broadening and covers a range of 600-1100 nm. The broadening of the spectrum of the rings is asymmetric and is directed mainly to the short-wavelength region of the spectrum. The annular structure of radiation in transverse direction of the beam is formed around individual filaments within the region of multiple filamentation, and at a distance of tens of meters from it begins to form a common ring structure surrounding post-filamentary channels. It is shown that post-filamentation channels at distances from the end of the filamentation zone, greatly exceeding the length of filamentation zone, contain the intensity sufficient for the formation of multiple filamentation in the optical elements and for excitation of the targets emission spectra.
DETERMINATION OF THE NONLINEAR REFRACTIVE INDEX OF CRYSTALLINE ZnSe BY MODIFIED METHOD OF Z-SCAN AT THZ FREQUENCIES.

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Recently, great progress has been made in the generation of high-intensity terahertz (THz) radiation, which has led to the opportunity to explore non-linear processes in the THz frequency range[1]. A key parameter characterizing the nonlinear response of a material is the non-linear refractive index, denoted as $n_2$. It has been predicted theoretically [2] that the THz nonlinear response of crystals is extremely large as a consequence of a strong vibrational contribution. An indirect assessment of $n_2$ for lithium niobate at THz frequencies was reported in [3], leading to a value of approximately 10^{-11}cm^2/W. In the present paper we report a direct experimental measurement of $n_2$ for crystalline ZnSe in the THz spectral range.

We used a femtosecond laser system (30 fs duration, pulse energy 2 mJ, repetition rate 1 kHz) as our radiation source. The radiation was split into two beams; one (pump) beam was used for generating the THz radiation and the other (probe) beam was used for monitoring the THz radiation. Generation of the THz radiation took place in the generator (TERA–AX; Avesta Project), which is based on phase-matched optical rectification in a lithium niobate crystal.

The THz pulse was focused and collimated using two lenses. The intensity of the radiation at the waist was 0.8×10^9 W/cm^2. The sample was translated through the beam waist region of the THz beam through use of a translation stage. The ZnSe sample was 300 m thick.

We measure the amplitude of the THz radiation by means of the electro-optical effect in a ZnTe crystal that is placed between the crossed polarizers. Then, as in the conventional Z-scan method, we translated the ZnSe crystal along the z axis. In distinction from the classical Z-scan method, we fix the value of the change of intensity of probe beam at the CCD camera in the area of maximum of the amplitude modulus of the THz pulse. Image processing with CCD-camera is performed as follows: we select an area in the vicinity of the resulting image. We sum the signal from these pixels, and we take this value to represent the amplitude modulus of the THz signal for this particular location in z.

Translation of the ZnSe crystal along the z-axis leads to a variation of the measured intensity that is characteristic of all Z-scan measurements. We thus use the standard formulas [4] for Z-scan measurements to estimate the non-linear refractive index.
We find $n^2 = 4.0 \pm 2.5 \times 10^{-11} \text{cm}^2/\text{W}$.

We measure an extremely large value of the $n^2$ coefficient of crystalline ZnSe in the THz spectral regime. This value is orders of magnitude larger than that of typical materials as measured at optical frequencies. Our results are consistent with the prediction of a recent [2] theoretical model that ascribes the origin of THz nonlinearities in crystals to a vibrational response that is orders of magnitude larger than typical electronic responses.

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HIGH-EFFICIENCY SPECTRAL SUPERCONTINUUM GENERATION IN THE TRANSPARENT NONLINEAR LIQUID JETS

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Using liquids as a nonlinear medium for SC generation allows to get broadband radiation with the higher intensity and shorter duration than, for example, in photonic crystal fibers, which makes it possible to increase the resolution in coherence tomography and pump-probe spectroscopy, to increase the information transmission distance through SC etc.

Method of high-efficiency SC generation in a liquid jet was proposed in paper [1] and was used in paper [2] as well. Since the nonlinear medium (liquid jet) was located before the pump radiation geometric center of the focus (caustic), the filament formation or breakdown occurred at higher density of pump intensity, increasing the conversion efficiency of SC. As a source of the femtosecond pulses a laser system Regulas 35F1K (Avesta Project) based on regenerative amplifier with the following parameters was used: pulse duration 30 fs, pulse energy up to 2.2 mJ, repetition rate 1 kHz and the center wavelength 800 nm. Femtosecond light was focused by a lens with a focal length 10 cm in a liquid jet with thickness 1.8 mm, which was located at 4 mm from the point of focus. The resulting spectrum of the SC was analyzed by spectrometers ASP-100 M (Avesta Project spectral range 200-1000 nm) and EPP-2000-NIR (StellarNet Inc, the spectral range of 900-1700 nm).

The hydrogen oxide, deuterium oxide and ethanol were used as liquids. Their parameters such as nonlinear refractive index, transparency window and other is presented in papers [1-4]. The SC
extending from 350 to 1400 nm in a hydrogen oxide jet, from 400 to 1600 nm in ethanol and from 350 to 1700 nm in deuterium oxide were obtained. Duration of SC temporal structures were 390 fs, 220 fs, and 250 fs respectively. For deuterium oxide and ethanol, filamentation threshold decreases in 1.5 times in comparison with hydrogen oxide. The SC spectrum generated in the hydrogen oxide had plateau-like shape in the range from 400 nm to 800 nm [1]. The SC spectrum generated in a deuterium oxide jet had clear defined components in the blue-green (350-550 nm) and infrared (900-1000 nm, 1300-1700 nm) regions [2]. The SC spectrum generated in an ethanol jet had pronounced components on the fundamental frequency and in the infrared region (900-1200 nm).

The maximum of conversion efficiency was observed in a deuterium oxide jet (70%), which is associated with a larger transparency window while SC generated in hydrogen oxide and ethanol had conversion efficiency of 50% and 60% respectively. Numerical simulation of SC formation was conducted in the LBullet software [5] to determine the intensity and duration of SC after propagation in the medium. It was shown that the intensity of SC generated in hydrogen oxide, ethanol and deuterium oxide have value of 0.8*1011 W/cm2, 4.7*1011 W/cm2 and 6.6*1011 W/cm² respectively.


EFFECTIVE H$_2$O IONIZATION IN AMBIENT AIR
AT 248 NM LASER WAVELENGTH


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Effects of water vapor on multiphoton ionization (MPI) of air by a 100 femtosecond KrF filament and narrowband KrF laser radiation were investigated. It was demonstrated that in both cases ionization of water becomes the dominant ionization process. For laboratory air (t= 20ºC, P= 1 atm and relative humidity $\eta = 40\%$ which corresponds to a water content of $\sim 1\%$) 2 different ionization regimes were observed depending on the ionizing species: resonance-enhanced multiphoton ionization (REMPI) and MPI. In the case of 100 fs KrF filament direct 3-photon MPI of water provided $\sim 90\%$ of total electron yield while the rest of the electrons in equal proportions were produced by (3+1) REMPI of N$_2$ and direct 3-photon MPI of O$_2$. For 25 ns KrF pulses electron yield due to (2+1) REMPI of water vapor containing in laboratory air was at least 2 orders of magnitude higher than in dry air where 3-photon MPI of O$_2$ took place.

Obtained data suggests that typical ion composition in UV induced moist air plasma including UV filaments consists mostly of H$_2$O$^+$, OH$^+$ ions and their derivatives. It requires review of both plasma decay and nonlinear propagation models.

NOVEL MECHANISM OF FEMTOSECOND PULSE SELF-SHORTENING BASED ON THE TRANSIENT REGIME OF MULTIPLE FILAMENTATION IN KERR MEDIA

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At present, the chirped pulse amplification (CPA) method has achieved a petawatt (1015 W) power level in solid-state femtosecond systems. As a rule, in such systems it is difficult to obtain pulses shorter than 20-30 fs because of narrowing of the spectrum during amplification. At the same time, for numerous applications in the strong fields – matter interaction and femtosecond optics (obtaining high-order harmonics and generating isolated attosecond pulses, investigating
the dynamics of ultrafast processes, etc.), it is important to obtain shorter pulses approaching a period of light wave. The solution to this problem is the development of nonlinear methods for pulse reduction at the output of femtosecond systems. The methods currently used are based on the broadening of the spectrum due to self-phase modulation (SPM) in media with Kerr nonlinearity with compensation of acquired phase modulation by using additional dispersive elements or without the use of these elements if the spectrally-limited pulses are formed directly in a nonlinear medium (self-compression). At the same time, currently developed self-compression methods are limited in energy by tens of mJ and do not have the prospect of further energy scaling.

In this paper, we present the results of investigations of a new mechanism for self-shortening of the duration of femtosecond pulses in thin optically transparent materials with Kerr nonlinearity. It is based on the formation of multiple filaments in the trailing edge of the pulse, which leads to a sharp increase in diffraction and refraction losses in the near-axis region. Experimental studies were carried out in a convergent Gaussian beam incident on a 1 mm thick fused silica plate. The duration of the original spectrally-limited pulse was 87 fs at a wavelength of 475 nm. The intensity of the radiation on the fused silica plate varied in the range of 0.8-2.9 TW/cm² in a beam 0.63 mm in diameter at 1/e² level. Using a diaphragm 100 μm in diameter, adjacent to the back surface of the sample, a region of uniform intensity in the central part of the beam was selected. The radiation at the exit of the diaphragm consisted of a central core whose size did not depend on the intensity in the initial beam, and conical emission with a divergence angle of up to 0.1 rad (the angle of convergence of the initial radiation was <0.01 rad). At intensity of 3 TW/cm², the radiation spectrum of the core had a shape close to Gaussian with a center at a wavelength of ~ 495 nm. The pulse duration was reduced to 19 fs.

The obtained experimental data show that the output pulse in the near-axis region is formed at the leading edge of the pulse, which is not subject to filamentation. At the same time, due to the SPM, the broadening and red shifting of the spectrum take place.

The mechanism of the observed phenomenon indicates that the method of self-shortening pulses based on transient regime of multiple filamentation allows its energy scaling.
5. Femtosecond radiation in spectroscopy and optical frequency metrology

Chair V N. Sorokin

FEMTOSECOND REFERENCE OSCILLATOR FOR CS/RB FOUNTAINS

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Compact and reliable microwave reference oscillators with \((1-5)\times10^{-15}\) (for average time \(\tau = 1 - 1000\) s) instability and low phase noise are needed for variety of applications – frequency etalons, based on atomic fountains, radio astronomy, radars, etc [1]. Common approach to their development is based on frequency stabilized lasers and further optical-to-microwave frequency division (OFD) [2]. Typically the laser is locked to high finesse Fabri – Perot (FP) optical cavity using Pound - Drever - Hall technique and an optical frequency comb based on a mode-locked femtosecond (fs) laser serves as a frequency divider. In our investigation we follow the above mentioned optical-to-microwave division scheme but as an optical reference we use the He-Ne/CH4 laser \((\lambda = 3.39\ \mu m)\) stabilized over narrow saturated dispersion resonances of CH4 spectral line.

This approach has some advantages. He-Ne/CH4 optical frequency standard (OFS) potentially has considerably less frequency drifts as compared to FP cavity fringes at average times \(\tau > 10s\). Also it is less sensitive to vibrations, temperature instability, Brownian fluctuations (in mirror surfaces, substrates and resonator body). Additionally the OFD scheme with a He-Ne/CH4 laser is relatively simple. It includes an optical frequency comb based on a fs Er fiber laser and difference frequency generation in a nonlinear crystal and only one parameter of the comb - pulse repetition rate - should be stabilized [3].

Our He-Ne/CH4 OFS is a compact zerodur block with high passive stability and low level of the spontaneous noise. It’s present instability is at the level \((5-7)\times10^{-15}\) \((\tau = 1 - 1000s)\) and further improvement to \((1-2)\times10^{-15}\) seems feasible [4].
At present work two reference oscillators based on a femtosecond Er fiber laser ($\lambda=1.55\mu$m) optical-to-microwave frequency divider and He-Ne/CH4 OFS were created. Preliminary measurements have demonstrated short term frequency instability of the output microwave signal at 1.5GHz at the level of $1\times10^{-14}$ ($\tau = 1$ s). Interrogative oscillator with such stability meets demands for application in Cs and Rb atomic fountains.

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References


TM-DOPED ALL-FIBER PASSIVELY MODE-LOCKED RING LASER FOR OPTICAL FREQUENCY STANDARDS IN MID-IR

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Using the mid-IR region in optical frequency standards, medical diagnostics, gas monitoring and other important applications is very perspective [1, 2]. The creation of cw tunable solid-state lasers based on crystals of II-VI compounds doped with divalent ions of transition metals [3] allows a new way for the future development of frequency-stabilized lasers and, consequently, compact optical frequency standards in mid-IR. One of such materials is ZnSe single crystals doped with Cr²⁺, which can be used for CH₄ two-modes saturation spectroscopy at 2.36 µm. This approach allows to reach Allan deviation at the level of 1·10⁻¹⁶ and push up frequency standards in mid-IR.

Optical frequency combs of mode-locked Er-doped fiber lasers have become an essential tool for aforesaid applications in near-infrared region [4], thus there’s a great necessity to find high-energy broadband sources of femtosecond pulses in mid-IR [5]. One of such potential sources is thulium-doped ultrashort-pulse fiber laser with broad gain spectrum from 1.7 to 2.1 µm, and a potential to produce high power and highly efficient tunable femtosecond pulses [6, 7].

We have made a first step towards a phase-coherent comb in mid-IR for compact optical frequency standard and demonstrate an all-fiber passively mode-locked ring laser based on normal dispersion active Tm-doped fiber. The laser generates stable 210 fs pulses with 26 nm spectral width at a center wavelength of 1940 nm with 150 mW output power and 11 MHz repetition rate.

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LOW-NOISE FIBER-BASED FEMTOSECOND FREQUENCY COMB

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We have developed Er-fiber based optical frequency comb referenced to a Hydrogen-maser. The main block of the frequency comb is femtosecond Er-fiber laser. Main output of the fiber laser is used for detection and stabilization of offset frequency. To obtain offset frequency signal output radiation of the fiber laser is amplified to average power of about 200 mW. Then supercontinuum with octave spectrum is obtained with highly-nonlinear fiber. The supercontinuum was used to detect offset frequency with a common-path f-2f interferometer. Detected offset frequency signal is locked to 10 MHz reference by current modulation of pump laser diode. Second output of the fiber laser was used for detection of pulse repetition rate. The pulse repetition rate was stabilized with a PZT-modulator and a PLL.

Spectrum of offset frequency in free-running regime is shown in Fig. 1(a). The signal-to-noise ratio of the offset frequency is more than 55 dB at 3 kHz resolution bandwidth. To estimate free-running linewidth of the offset frequency a weak lock of the offset frequency to 10MHz reference was used. In the weak lock regime (without spectrum narrowing of the offset frequency) we obtained minimum offset frequency linewidth of about 4kHz at 2kHz resolution bandwidth (Fig. 1(b)). To the best of our knowledge the value is one of the narrowest offset frequency linewidths for fiber frequency combs. The narrow spectrum of the offset frequency suggests low phase noise of pulse repetition rate signal and low comb-line frequency noise of our frequency comb.

Fig. 1(a). Spectrum of offset frequency in free-running regime (3 kHz resolution bandwidth).

Fig. 1(b). Spectrum of offset frequency in weak lock regime (2 kHz resolution bandwidth).

Also we measured phase noise of pulse repetition rate (third harmonic of pulse repetition rate, 197.6 MHz) and compared the phase noise with one of a commercial fiber frequency comb (first
The results of the measurements are shown in Fig. 2. Black line corresponds to phase noise of free-running commercial frequency comb, red line indicates phase noise of commercial frequency comb locked to an ultrastable optical reference and blue line represents phase noise of our free-running frequency comb. We can see that in free-running regime the phase noise of our frequency comb is significantly less than one of the commercial frequency comb. And the phase noise of our free-running frequency comb is comparable to the phase noise of the commercial frequency comb locked to an ultrastable optical reference.

Fig. 2. Power spectral density of the phase noise of tested frequency combs.
ERBIUM DOPED FIBER LASER BASED COMB-GENERATORS WITH THE FREQUENCY PARAMETERS STABILIZATION

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Extremely high-stable fiber laser based optical comb-generators are one of the most convenient and reliable solution for a wide range of tasks concerning to a high precision frequency measurement, synchronization etc [1, 2]. The most important part of the fiber laser based comb-generator design is to provide high-accurate and effective internal subsystems and construction features which control and stabilize all own frequency parameters of the comb-generator [1]. We would like to represent some results of the fiber laser based comb-generator development, which have been obtained during the manufacturing comb-generator systems by our company Avesta Ltd. with a collaboration with the Lebedev Physical Institute of the Russian academy of sciences and the Researching Institute of the Physical Technical and Radio Technical Measurements (VNIIFTRI).

In general, there are two frequency parameters, which have to be strongly stabilized for obtaining the proper output signal parameters accuracy: a carrier-envelope offset frequency (f_{ceo}) and a repetition rate (f_{rep}) [3]. The following method has been used for the f_{ceo} stabilization: the f_{ceo} value has been detected from a beat-note signal by an f2f interferometer and processed by a Phase Lock Loop (PLL) unit, and then a control signal from PLL unit has been gone into a master-oscillator pump control unit for neat and smooth correction of the master-oscillator pump power which leads to the f_{ceo} value stabilization [4]. For the f_{rep} stabilization the bet-note signal has been obtained from a mixing of the master-oscillator output signal and a high-stable external frequency reference, then it has been processed by the PLL unit, which was forming the control signal for three independent intracavity stabilization subsystems (thermoelectrical controller, piezo-motor controller and electro-optical modulator) for proper and stable locking of the f_{rep} value [4].

During this work, two different tasks was solved. The first one is the erbium doped fiber laser based comb-generator development with the f_{ceo} and the f_{rep} stabilization where f_{rep} has been stabilized by the mixing of the master-oscillator output signal with the reference based on a semiconductor diode laser stabilized by an ULE resonator. The second one is the optical clock with $10^{-14} – 10^{-15}$ stability (averaging time 1 s) development where the frequency parameters
stabilization has been realized by using a method of a difference frequency generation (DFG) [6] and the mixing with 3,39 nm wavelength reference based on a methane-stabilized HeNe laser.

As a result, we represent commercially available, reliable and complete fiber laser based comb-generator systems with frequency parameters stabilization values described below. For the first task: concerning the $f_{\text{rep}}$ we achieved RMS of $f_{\text{rep}} = 0,2$ mHz, $\Delta f_{\text{rep}}/f_{\text{rep}} = 3,1\cdot10^{-12}$ at gate time = 1 s; concerning the $f_{\text{ceo}}$ we achieved RMS of $f_{\text{ceo}} = 1$ mHz, $\Delta f_{\text{ceo}}/f_{\text{ceo}} = 1\cdot10^{-10}$ at gate time = 1 s; total relative error of the system is equal to $0,24\cdot10^{-17}$. For the second task: we achieved RMS of $f_{\text{rep}} = 0,02$ mHz and total relative instability of the system about $3\cdot10^{-13}$, besides, in the case of using the 26-s harmonic of the $f_{\text{rep}}$ we get RMS of 26-s harmonic of the $f_{\text{rep}} = 0,01$ mHz and it’s relative instability (averaging time 1 s) approx. $1\cdot10^{-14}$.


FEMTOSECOND PULSES AND KERR COMBS IN MICRORESONATORS.

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The advent of microresonator based Kerr combs paves a path to novel applications where traditional optical frequency combs requiring bulky apparatus cannot be used. In this case, frequency comb is formed spontaneously in optical ring-type or whispering gallery microresonator in four-wave mixing cascaded processes. Though initial expectations were somewhat mitigated by intrinsic chaotic character of first generation combs, it was shown that coherent mode-locked combs which are associated with self-supporting femtosecond optical pulses (solitons) are still possible without significant additional efforts on different platforms.

Recent theoretical and experimental results in soliton combs from the Russian Quantum Center are reported. Key advantages of microresonator based frequency combs are their compact form factor, high power per comb line, and ability to access microwave repetition rates, relevant for many application including high capacity telecommunications or microwave photonics. It was also revealed that coherent Kerr combs are possible not only for anomalous group velocity dispersion necessary for bright optical solitons but also in normal dispersion systems using the so-called “platicons” – solitonic like flat-topped waveforms. This opens the ability to generate coherent combs in the UV or mid IR spectral range with the gain bandwidth limited only by the transparency window. Recently demonstrated microresonator based dual frequency combs open the possibility to transfer optical spectral measurements to radio-frequency domain. New experimental methods of Kerr soliton generation using cheap regular wide spectrum multifrequency diode laser self-injection-locked high-Q microresonator will be discussed.