COFIL 2024

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International Conference on

Laser Filamentation

25 - 29 August 2024 **Tianjin, China**

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Brief Introduction of COFIL

The International Conference on Laser filamentation (COFIL) is a traditional conference held every two years. The COFIL 2024 will be a continuation of a series of successful conferences in Canada (2006 and 2016), France (2008), USA (2012), China (2014), Switzerland (2018), Greece (2010 and 2022). The topics of the conference focus on novel phenomena and techniques for the studies of filamentation and related applications. The COFIL 2024 is held during 25 - 29 August, 2024, at Tianjin Crystal Palace Hotel (天津水晶宫酒店). This meeting is organized by the Institute of Modern Optics, Nankai University, in cooperation with international and domestic scientific institutions.

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Location:

Conference Agenda

Sightseeing spot in Tianjin

Tianjin ancient cultural street

The porcelain house

Tianjin Cultural Centre

Monday, August 26, 2024

(Location: Crystal Ballroom, 3rd floor of Tianjin crystal hotel)

Presider: Prof. Weiwei Liu, Nankai University

Abstract

Lightning in a Forest (Wild) Fire: Mechanism at the Molecular Level[1]

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The mechanism of lightning induced forest fires in countries/regions of high latitude can be explained qualitatively based upon the mechanism of lightning at the molecular level recently proposed by the authors and co-workers^[1]. The mechanism is based on two phenomena, namely, internal charge separation inside the atmospheric cloud particles and the existence of a layer of positively charged hydrogen atoms sticking out of the surface of the liquid layer of water on the surface of rimers. Strong turbulence-driven collisions of the ice particles and water droplets with the rimers break up the ice particles and water droplets into positively and negatively charged fragments leading to charge separation. Hot weather in a forest contributes to the updraft of hot and less humid air. Therefore, lightning would have a high probability of lighting up and burning the dry biological materials in the ground of the forest, leading to a forest (wild) fire. Once the burning of trees and other plants occurs, it releases a lot of heat and moisture together with a lot of smoke particles (aerosols) becoming a strong updraft. The condition for creating a lightning is again satisfied which would result in further lightning high above the forest wild fire. Such events could result in the induction of lightning over neighbouring forests as well as distant forests in the northern colder part of a large region. An example is western Canada during the month of August, 2023. Forest wild fire spread and "jumped" locally and new but delayed forest fires spread over large regions. Delayed forest fire took place even in Yellowknife in the NWT. The latter might have been induced by the forest fire in the south. To reduce the probability of lightning in a forest, the weather should be cold so as to reduce the rate of moisture evaporation and the strength of the updraft. Unfortunately, climate change seems to push the weather towards a warmer one. Lightning and forest fires are thus unavoidable and will intensify in the future because the weather is getting warmer. Satellite and laser technologies might help in reducing the spread of forest fire. More work in this direction might have to be carried out in the future.

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Evidence of optical amplification without population inversion of N_2^+ **using ultrafast XUV spectroscopy**

Rostyslav Danylo1 , Guillaume Lambert1 , Mykyta Redkin2 , Aurélien Houard1 , Xiang Zhang3 , Liang Xu3, Yi Liu3, Arnaud Couairon4, Vladimir Tikhonchuk5 and André Mysyrowicz1*

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We show that a very strong gain is obtained at 391.4 nm without requiring a population inversion between initial and final levels of the transition. The amplifying medium is N_2^+ at low density, pumped by an intense laser source at a longer wavelength, i.e. 800 nm. It is probed by measuring with femtosecond precision the transmission of XUV high harmonics through the gas, displaying the evolution of the relevant electronic levels during the amplification process. We demonstrate that a third level, forming a "V scheme" with the initial and final levels, is involved in this case.

Second harmonic emission in dual-laser-induced air plasma

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We report a surprising strong second harmonic generation from a dual-laserinduced air plasm[a\[1\]](#page-20-0)[\[2\]](#page-20-1)[\[3\].](#page-20-2) Several discovered features are not satisfactorily accounted for with previously proposed models. The efficiency of the second harmonic process demonstrates a counterintuitive resilience against changes in fundamental pulse duration. The optical polarization of the second harmonic emission also does not simply co-rotate with that of either fundamental beam, as opposed to the single-laser counterpart process in air. We measure the emission properties of such nonlinear process, itemize the tunable parameters, and analyse the polarimetric relationship between fundamental and second harmonic components. These experimental studies will aid the sifting and development of a theoretical basis that explains the observed features of the nonlinear interaction.

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Terahertz emissions from foil targets irradiated by ultraintense laser pulses

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Intense sources of terahertz radiation are attracting more and more interest for various applications^[1]. A current challenge is nowadays to produce broadband THz pulses with mJ-level energies. An auspicious approach for this purpose is to irradiate solid targets at relativistic laser intensities > 10^{18} W/cm². In this regime, coherent transition radiation (CTR) from electron bunches exiting the rear plasma boundary in gases can lead to intense THz emissions, characterized by a few-100-μJ energy yield and > 10 GV/m field strength^[2]. Recently, experimental measurements^[3] reported an efficient production of terawatt-scale, mJ-level THz pulses, from high-intensity picosecond laser beams irradiating metal foils. CTR from relativistic laser-solid interactions benefits from a stronger absorption of the laser energy into MeV-range electrons. Yet, owing to its high density ($\sim 10^{19-21}$ cm⁻³), the hot-electron population does not only radiate via CTR when exiting a solid foil. Less energetic electrons actually get reflected in the strong charge-separation field that they set up in vacuum. This results in an additional coherent, synchrotron-type radiation of polarity opposite to that of CTR. Moreover, the sheath electric field induced by the hot electrons on both sides of the target subsequently sets into motion the surface ions over picosecond timescales and lead to a dipole-type, low-frequency radiation able to contribute to the THz spectrum.

In the present talk, using two-dimensional particle-in-cell simulations we first investigate the mechanisms of terahertz emissions in sub-micrometer-thick solid foils driven by ultraintense ($\sim 10^{20}$ W/cm²), ultrashort (30 fs) laser pulses at normal incidence. The range of target thicknesses extends from 0.5 μm down to the relativistic transparency regime (~ 15 nm) that optimizes fs laser-driven ion acceleration. By disentangling the fields emitted by longitudinal and transverse currents, our analysis reveals that, within the first picosecond after the interaction, THz emission occurs in bursts as a result of coherent transition radiation by the recirculating hot electrons and antenna-type emission by the shielding electron currents traveling along the fastexpanding target surfaces^[3].

Next, we theoretically focus on the radiation emitted by the energetic electrons exiting the backside of a solid target. Our model takes account of the coherent transition radiation due to electrons crossing the plasma-vacuum interface as well as of the synchrotron radiation due to their deflection and deceleration in the sheath field set up near the target backside. After showing that both mechanisms tend to largely compensate each other when all the electrons are pulled back into the target, we then demonstrate the sensitivity of this radiation to a percent-level fraction of escaping electrons. The same sheath field that confines most of the fast electrons around the target rapidly sets into motion the surface ions. We describe the THz emission from these accelerated ions and their accompanying hot electrons by means of a plasma expansion model that allows for finite foil size and multidimensional effects. Under conditions typical of current ultrashort laser-solid experiments, we find that the THz radiation from the expanding plasma is much less energetic—by one to three orders of magnitude—than that due to the early-time motion of the fast electrons^[5].

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Filamentation enabled cryptography and THz metamaterial-based ultrafast modulators

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Laser filamentation is nowadays an enabling technology for applications across a very broad spectrum, from nonlinear THz science to materials engineering, and even lightning control.

Here we will be reviewing some of our recent advances in the field. We will show how optical information encoded in holograms is transferred by means of ultrashort laser filaments propagating in highly nonlinear and turbulent media. The initial optical information is completely scrambled and cannot be retrieved by any experimental or physical modeling system. Yet, we demonstrate that neural networks trained on experimental data, provide a robust way to fully recover the original hologram images^[1]. This advance opens the way to applications in secure free space optical communication links and cryptography.

We will also discuss our progress in developing 3-dimensioal (3D) chiral metamaterials that present strong broadband optical activity without ellipticity^[2]. The design of the 3D chiral metamaterials consists of pairs of U-shaped resonators that are "twisted" vertically and present a large and ultrabroadband pure optical activity in the low THz regime. The 3D chiral metamaterial structure was fabricated though Direct Laser Writing by Multiphoton Polymerization, followed by selective metallization through electroless plating with silver, and characterized through THz time-domain spectroscopy.

Finally, we will present the development of ultrafast THz modulators based on cavities with graphene as the active medium $^{[3]}$. A thin film graphene-based THz perfect absorbing device whose absorption and phase characteristics can be modulated through THz self-actions in the subpicosecond time scale. The device consists of a single-layer graphene placed on an ionic liquid substrate, back-plated by a metallic back-reflector, with the graphene doping level mediated through electrostatic gating. We experimentally record an absorption modulation of more than 3 orders of magnitude from the initial perfect absorption state, when the device is illuminated with THz field strengths in the range of 102 to 654 kV/cm.

Acknowledgements

This work has been partially funded by the Hellenic Foundation for Research and Innovation (H.F.R.I.) under the "2nd Call for H.F.R.I. Research Projects to support Faculty members and Researchers" (Project Number: 4542).

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Vortex and vector lasing of nitrogen ions

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Cavity-free lasing of nitrogen ions has attracted many attentions, not only due to the rich physics involved, but also due to its unique potential to create a virtual laser source in the sky $^{[1]}$. In this talk, I will discuss our recent progress on vortex and vectorial lasing of nitrogen ions^[2,3]. We first demonstrated that a vortex seed pulse injected into the nitrogen gas plasma excited by a Gaussian pulse is amplified substantially and its OAM property can be maintained. Second, we show that a vortex superfluorescent radiation at 391.4 nm carrying the same photon orbital angular momentum as the pump beam is obtained by focusing a vortex pump beam at 800 nm in N_2 gas. With the injection of a Gaussian seed beam at 391 nm, the radiation is amplified with the vorticity unchanged. Finally, we employed a pump beam with a cylindrical vector mode, the Gaussian seed beam is correspondingly amplified into a cylindrical vector beam. The underlying physics was attributed to the anisotropic optical gain of the plasma media due to the permanent alignment of the nitrogen ions $\tilde{A}^{[4]}$. The above results open the interesting perspective of OAM beam amplification and manipulation in the ambient air, which is of interest for free-space optical communication, remote sensing, and quantum information relay in atmosphere.

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Laser ignition of fuel-lean mixtures by femtosecond filmamentation pulses

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Laser ignition (LI) allows for precise manipulation of ignition timing and location and is a promising electrode-less alternative to electronic spark ignition of lean-fuel/air mixtures, offering high thermal efficiency with low harmful emissions. One of the most widely adopted LI methods is anosecond laser-induced spark ignition, which creates a plasma spark in the combustible mixture through breakdown of fuel molecules, producing localized temperatures of $10^5 \sim 10^6$ K. On the other hand, it is commonly deemed that igniting lean-fuel mixtures by an ultrashort femtosecond (fs) laser is hard to realize, since avalanche breakdown cannot occur on the fs timescale, and the fslaserinduced plasma temperature is 1–2 orders of magnitude smaller than that pumped by ns lasers. In this contribution, we present the experimental realization of igniting a lean methane/air mixture (see Fig. 1) by an ultrashort femtosecond filamentation pulse^[1-5]. It is discovered that the fs laser filamentation can induce 100% success rate of fs-LI with ultralow sub-mJ minimum ignition energy, exhibiting distinct contrast to the general understanding that it is hard to achieve fs- $LI^{[1-3]}$. We show that the minimum ignition energies depend on filamentation formation, and find that the fs-LI becomes easier in a Goldilocks focal zone, where a crucial balance between the length of igniting

"line" kernel and the plasma density of the fs laser filament is achieved $[2]$. We then show that the so-called Goldilocks focal zone can be broken through by using a telescopic filament $[4]$, and propose that fs-LI can be ascribed to a novel non-resonant photochemical ignition mechanism^[5], in which the multiphoton/tunnel ionization of the lean-fuel mixture by the ultraintense femtosecond laser pulse produces the reactive radicals through various dissociation and chain reaction pathways, and thus result in the successful ignition at the micro-joule level.

Figure 1. Dynamical evolution of the flame ignited by femtosecond laser filamentation pulses $^{[2][2]}$.

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Photoelectron emission and high-order harmonic generation in solids excited by strong femtosecond laser pulses

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The interaction between strong femtosecond laser and matter leads to well-known strong-field phenomena, such as tunneling ionization and high-order harmonic generation. These studies not only reveal the mechanism of light-matter interaction but also provide methods for generating advanced electron and light sources on a benchtop scale. In recent years, our study of strong-field physics has expanded from gas to solid materials. Here, we present our recent progress in the field of photoelectron emission and high-order harmonic generation in solids. In the case of photoelectron emission, we have successfully generated an ultrashort ultraviolet extreme light source with high flux through gaseous high-order harmonics and developed an ultrafast extreme ultraviolet photoelectron emission microscope^[1]. Using this advanced microscope with ultra-high time, space, and energy resolution, we have demonstrated the characterization and manipulation of electron pulses emitted from nanostructures on an ultrafast timescale^{[2-1}] ^{4]}. Regarding high-order harmonic generation in solids, we have determined the magnitude of electron dephasing time $[5]$ and explored the dynamics of photoexcited e lectrons^[6-8]. Furthermore, we have demonstrated the manipulation of two carrier dynamic processes driven by light waves in zinc oxide crystals $[6]$. One process involves electron-hole collision recombination leading to high-order harmonic generation, while the other involves electron-electron impact excitation leading to stimulated simulation. We have effectively switched between these two dynamic processes using a weak control laser. This all-optical switch exhibits ultrafast speed, low threshold, and broad spectral responsiveness.

Figure 1. Ultrafast extreme ultraviolet photoemission electron microscope based on gaseous high-order harmonic generation^[1].

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Tailoring femtosecond filaments aerodynamics for applications

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Through optical ionization, a laser pulse undergoing filamentation in air deposit energy in the gas, resulting in the formation of a thin column of hot gas and by the emission of a radial pressure wave^[1-2]. These aerodynamics effects of filamentation can be exploited for many applications such as the guiding of electric discharge^[3], the formation of optical air waveguide^[4], the clearing of clouds for optical transmission ^[5], or the control of supersonic vehicles^[6]. Because filament induced aerodynamics occur on a relatively long timescale ranging from the microsecond to several milliseconds, they can be strongly enhanced by the accumulation of laser pulses at high repetition rate.

In this talk I will review recent experiments performed at LOA, where filament aerodynamics has been tailored using structured beams to organize multiple filamentation $[6-7]$, and where the cumulative effect has been used to enhance these aerodynamics effects $[8-9]$.

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Femtosecond Filamentation with THz-Frequency Pulse Bursts

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The generation of plasma by ultrashort laser pulses is a fundament for many scientific and industrial applications, such as micromachining and materials processing^[1], laser-induced breakdown spectroscopy^[2], or pulsed laser deposition^[3]. Plasma generation at powers exceeding 1 GW/cm² is complex, influenced by self-phase modulation, Kerr lensing, and nonlinear absorption. Under strong-focusing conditions with a single pulse, plasma shielding in the prefocal area limits the fluence that can be delivered to the focus at higher pulse energies. This stays in contrast to burst-mode operation, where the total energy can be increased by adding more pulses to the burst instead of increasing the peak intensity (See Fig. 1).

Figure 1. Luminescence curves derived from the recorded plasma distributions (see inlays) a) with a single 220 fs pulse of various energies. Inlay: 1.2 mJ. b) with an energy of 400 μJ per pulse and a varying number of pulses. In burst mode (> 1 pulse), the result for a pulse spacing of 2 ps is shown. Inlay: 5-pulses.

We developed a mode-locked oscillator-amplifier system exploiting the Vernier effect to generate multi-millijoule bursts of ultrashort pulses $[4,5]$. Unlike conventional burst formation methods, our approach maintains identical spatial properties of all pulses. Here, we demonstrate the generation of millijoule Vernier bursts of 210 fs pulses with tunable picosecond spacing, capable of inducing plasma in air. To generate the plasma in air, we focus the optical beam with a diameter of about 2.6 mm (FWHM) with an $f = 10$ cm focusing lens, to provide hard focusing that dominates the setting of the focal position over the Kerr effect in air. The beam diameter at the focus was measured to be about 30 μm (FWHM). The generated plasma is monitored by a camera that we use to measure the plasma luminescence in combination with a magnification lens. As can be seen in Fig. 1a), an increase in the pulse energy does not lead to a peak luminescence increase, because of plasma generated in the prefocal region. In contrast, when applying a burst of picosecond-spaced pulses (Fig. 1b), we see that by keeping the pulse energy constant while increasing the number of pulses, an increase of peak luminescence by more than 40% is observed.

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THz emission from long DC-biased single colour femtosecond filaments

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THz generation efficiency enhanced greatly if femtosecond single colour single filament is biased by a DC field with field strength near the breakdown of a medium [1]. Angular and energy spectra depend on the DC field strength as well. Normally the length of DC electrodes along the optical axes is much shorter than the filament length. Besides THz energy is limited if a single filament regime is used. To overcome these limitations and enhance THz production efficiency further we propose longer electrodes with the length comparable to the filament length and multifilamentation regime with higher laser pulse energy. We utilize both stochastic and regularised multifilament using amplitude or phase wavefront modulation of the initial femtosecond beam.

The TW Ti:Sa femtosecond laser (50 mJ, 50 fs, 10 Hz) was used. Amplitude or phase masks were placed just before the focusing lens (focal length from 1 to 10 m). Filament length and position as well as energy density inside it were measured using wideband acoustic method [2]. The electrode length was changed to fit the filament length. Transverse image of energy flow in the filament was obtained slicing the filament cross section by a wedge. The applied DC field was close to the breakdown. THz emission was measured using Goley cell or bolometer. THz spectrum was deduced from autocorrelation measurements with Michelson interferometer.

We showed that with long electrodes THz signal are added constructively while the angular distribution is getting narrower. The optimum length equals roughly to the filament length deduced from acoustic measurements [3]. We also achieved pronounced enhancement in the THz yield with laser pulse energies (peak powers) corresponding to the multifilamentation regime. The paper will also present experimental and numerical data with loosely focused regularized beams and beam undergone selffocusing passing a long atmospheric path.

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Intense terahertz wave generation from air plasma induced by femtosecond laser

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Terahertz (THz) sources generated using an air-excited plasma have been extensively studied because of their high performance and extensive applications in THz spectroscopy and imaging and THz–matter interactions. In 2000, Cook et al. demonstrated strong THz waves generated from air plasma excited using two-color pulses of fundamental and its second harmonic (SH) waves and presented a four-wave mixing model to explain the generation. Since then, extensive studies have been carried out on the physical mechanism and approaches for an enhancement of the THz generation from two-color fields by manipulating the transverse asymmetry of the electric field in the plasma to obtain an advanced and practical method to produce intense THz waves. Among the models, the photocurrent model proposed by Kim et al. has been widely applied and further developed.

We propose a THz emission system using three-color lasers with adjustable nearinfrared wavelengths obtained using an OPA, its SH, and an additional 800 nmultrafast pulse. We demonstrate a large stable THz enhancement of the near-infrared two-color fields after the addition of the 800 nm laser. Moreover, we obtained the variation trends of the THz strengthening effect with important parameters in the system such as the polarizations and energies of the lasers, and then analyzed the optimal conditions for the maximum THz enhancement. The modulation of the THz spectral energy distribution and central frequency shifting after the addition of an 800 nm pulse could provide valuable insights for the generation of THz radiations with specific frequency distributions for certain purposes.

Figure.1 Experimental schematic diagram. OPA is optical parametric amplifier. PM1, PM2, PM3 are off-axis parabolic mirrors.

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High-repetition rate femtosecond laser air filamentation: challenges and opportunities

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With the rapid development of intense femtosecond laser technology at kHz and even 100kHz high repetition rate, the high repetition rate femtosecond laser induced filamentation in air provides unprecedented opportunities for laser machining, laser telecommunication through fog, laser-induced condensation and snow formation, laser lightning control and etc. Due to the photothermal relaxation process of air molecules in the millisecond time scale, the pulse cumulative effects of high-repetition-rate intense femtosecond laser on filamentation in air will inevitably occur. Deeply understanding the influence of pulse cumulative effects on the high-repetition-rate laser filamentation process is the key to further explore the new applications of laser air filament. In this talk, we shall focus on the above key issues and present our recent activities on high repetition rate laser filamentation as well as its applications [1-7].

The research activities were in part supported by NSAF joint fund (No. U2130123), International Partnership Program of the Chinese Academy of Sciences (No. 181231KYSB20200033) and Shanghai Science and Technology Program (No. 21511105000).

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1D/3D MAXWELL-BLOCH modelling of nitrogen lasing from plasma filaments

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The amplification of UV radiation in plasma filaments is a multiscale (from the femtosecond timescale of the intense IR pulse that creates the filament to the picosecond scale of amplification dynamics and the nanosecond scale of plasma hydrodynamics) and multiphysics (plasma physics, electronic collisions, laser-plasma interaction) problem.

In this talk we will present the computational framework developed at tha IFN-GV to model lasing from N2 and N₂⁺ plasma filaments, 1D-DeepOne^[1] and 3D Dagon^[2], and some results obtained with them, when combined with experimental results: unveiling the lasing mechanism in neutral nitrogen filaments^[3,4,5], electron collisional excitation, and suggesting that the wavefront of the amplified UV beam carries information about the spatio-temporal dynamics of the plasma amplifier^[6]. Our modelling results related to N_2^+ lasing^[7-8] also provided hints about the amplification dynamics. Finally, we will show that the similarities with plasma-based seeded soft Xray lasers allows to transpose well known techniques of this field onto nitrogen lasing, allowing the optimization of these amplifiers.

Figure 1. (upper panel) Adiabatic and (lower panel) fully time-dependent modelling of the amplification of a UV beam through a nitrogen filament several centimetres long. The figure shows the intensity of an amplified UV seed through more than 3 cm of filament plasma.

This work has received the support of a Beca Leonardo a Investigadores y Creadores Culturales 2021 de la Fundación BBVA and the Ministerio de Ciencia e Innovación MCIN/AEI/10.13039/ 501100011033 and European Union « NextGenerationEU »/PRTR grant CNS2023-144058

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Filamentary propagation and modification characteristics induced by picosecond laser in sapphire

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The paper firstly studies the propagation behavior and modification characteristics of filament in sapphire excited by picosecond laser (ps) single pulse. The self-designed and built instantaneous acquisition method and system of filamentary plasma are used to study the effects of laser peak power and geometric focusing position on filamentary propagation and modification. Based on the simulation and analysis of the temporal and spatial distribution of ps single pulse and plasma density during filamentary propagation, the different dynamic equilibrium and main ionization mechanisms in various stages are revealed. The modification type of sapphire trace affected by filamentation of ps single pulse is determined and the modification mechanism is analyzed. A ceramic polycrystalline region is formed in the filamentary trace. The phase is transited from α -Al₂O₃ to γ -Al₂O₃, which provides a high possibility for the filamentinduced laser high-precision manufacturing of sapphire. Further, the optimization filamentary propagation in sapphire excited by the burst mode modulated by the MHz level repetition rate sub-pulse is proposed and studied. Uniform diameter $(\sim 2.5 \text{ }\mu\text{m})$ filamentary trace with a maximum aspect ratio of 273.3 is achieved in sapphire.

Figure 1 Overall morphology of the filamentary plasma induced by burst mode with *B*3F in sapphire.

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Millijoule terahertz radiation from laser wakefields in non-uniform plasmas

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Terahertz (THz) radiation with energy up to the millijoule level is essential for applications in THz nonlinear optics, electron acceleration and reshaping, and molecular dynamics. We report the experimental measurement of millijoule THz radiation emitted in the backward direction from laser wakefields driven by a femtosecond laser pulse of few joules interacting with a gas target. By utilizing frequency-resolved energy measurement, it is found that the THz spectrum exhibits two peaks located at about 4.5 THz and 9.0 THz, respectively. In particular, the high frequency component emerges when the drive laser energy exceeds 1.26 J, at which electron acceleration in the forward direction is detected simultaneously. Theoretical analysis and particle-in-cell simulations indicate that the THz radiation is generated via mode conversion from the laser wakefields excited in plasma with an up-ramp profile, where radiations both at the local electron plasma frequency and its harmonics are produced. Such intense THz sources may find many applications in ultrafast science, e.g., manipulating the transient states of matter.

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Characterization of self-focusing of femtosecond laser pulses in optical media

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With the increase in the input power of the femtosecond laser pulse, the pulse will successively undergo several processes in optical media, such as from linear propagation to self-focusing and then to filamentation. The transition from the linear to nonlinear focusing regime is commonly used to determine the critical power for selffocusing (P_{cr}) of the laser pulse. The determination of the critical power is crucial for understanding the femtosecond laser filamentation mechanisms and for predicting filament distribution and evolution, thereby facilitating filamentation applications. There are several approaches to investigate the $P_{cr}^{[1-4]}$. In this presentation, we will introduce two methods, namely the fluorescence and S-scan methods, for studying the self-focusing behavior and determining the P_{cr} of the femtosecond Gaussian and vortex laser pulses $[5,6]$.

Figure 1. (a-d) Fluorescence and (e-h) S-scan method to study the self-focusing behavior and P_{cr} of femtosecond Gaussian and vortex pulses in air and fused silica, respectively.

First, by measuring the 337 nm fluorescence emission from the air ionization region using a PMT, as shown in Figs. 1(a-d), the signal does not change too much when the laser energy is relatively low, and has a rapid increase with a further increase in the energy, which is similar to that obtained by using the focus-shifting method. The critical powers of femtosecond Gaussian and vortex beams are successfully obtained. The second method is based on the evaluation of the spectral broadening of the laser pulse with different energies. The rapid broadening and saturation of the spectrum indicate the self-focusing process and intensity clamping effect in the laser pulse, respectively, as shown in Figs. 1(f-h). Eventually, the linear, self-focusing and mature filamentation regimes for Gaussian and vortex femtosecond pulses in fused silica are successfully distinguished, and the corresponding critical powers are obtained. Both methods are suitable for determining the critical power for self-focusing of both femtosecond Gaussian and vortex lasers.

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Self-focused pulse propagation is mediated by spatiotemporal optical vortices

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The dynamics of high-intensity laser pulses undergoing self-focused propagation in a nonlinear medium can be understood in terms of the topological constraints imposed by the formation and evolution of spatiotemporal optical vortices (STOVs) [1- 5]. I will first review STOVs and then show their role in self-focused propagation and filamentation. During self-focusing pulse collapse, STOVs are born from point phase defects on the sides of the pulse nucleated by spatiotemporal phase shear. These defects grow into closed loops of spatiotemporal vorticity that initially exclude the pulse propagation axis, but then reconnect to form a pair of toroidal vortex rings that wrap around it. STOVs constrain the intrapulse flow of electromagnetic energy, controlling the focusing-defocusing cycles and pulse splitting inherent to nonlinear pulse propagation. We illustrate this in two widely studied but very different regimes, relativistic self-focusing in plasma and non-relativistic self-focusing in gas, demonstrating that STOVs mediate nonlinear propagation irrespective of the detailed physics.

Figure 1. (a)-(h): Simulation [1,5] of a nonrelativistic Gaussian laser pulse (25 mJ, 500 fs FWHM, $\lambda = 0.8 \mu m$, $w_0 = 1 \mu m$, $P/P_{cr} = 5$) self-focusing into a semi-infinite argon gas, density 2.5×10^{19} cm⁻³. Red arrow = propagation direction. Pulse splitting is driven by STOV evolution. (i)-(i): STOV nucleation and evolution.

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Bidirectional cascaded superfluorescent lasing in air enabled by resonant third harmonic photon exchange from nitrogen to argon

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Cavity-free lasing in atmospheric air has stimulated intense research towards fundamental understanding of underlying physical mechanisms. Here we identify a new mechanism -- third harmonic photon mediated resonant energy transfer pathway leading to population inversion in argon via initial three-photon excitation of nitrogen molecules irradiated by intense 261 nm pulses -- that enables bidirectional two-color cascaded lasing in atmospheric air [1]. By making pump-probe measurements, we conclusively show that such cascaded lasing results from superfluorescence (SF) rather than amplified spontaneous emission (ASE). Such cascaded lasing with the capability of producing bidirectional multicolor coherent pulses opens additional possibilities for remote sensing applications.

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Tailoring and stabilization of femtosecond filaments in spectral and spatial domains via laser beam modulation

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Femtosecond laser filaments, which formation and evolution is intrinsically nonlinear phenomenon, are challenging objects to take their parameters under control. Moreover, a separate challenge is to make these parameters pulse-to-pulse reproducible and robust against initial laser beam fluctuations.

In our report, we discuss the opportunities that amplitude and phase modulation of the beam open up in terms of management and stabilization of supercontinuum after filamentation and its angular structure, filaments positions in multifilament arrays and energy deposition into the medium. For this purpose, we applied a set of complementary experimental techniques to obtain diverse data: single-shot anglewavelength spectra, transverse Fourier patterns of radiation after filamentation, beam profiles and acoustic signals from filaments. For each propagation regime, a sample of about $10^2 - 10^3$ laser shots is analyzed providing data on pulse-to-pulse stability of the parameters under investigation. The analysis is made for the pulse power ranging from below 1 critical power for self-focusing (P_{cr}) up to 40 P_{cr} , so that transition from singlefilament to multifilament regime and its effect on filament parameters and their stability is traced.

Figure 1. (1,2): Spectral position of the brightest red-shifted hump in the filament spectrum, $(3,4)$: energy contained in this spectral maximum. $(1,3)$: Initial beam is modulated by 4-hole amplitude mask. (2,4): the beam without modulation. Each point on the graphs corresponds to a single laser shot.

The main results include:

1. Stabilization of the supercontinuum brightest red-shifted maximum in spatial, spectral and energy domains (see fig.1 for the spectral and energy data) via beam amplitude modulation [1].

2. Beam amplitude modulation allows to radically change the picture of conical emission (CE): modulation by a 4-hole mask suppresses CE in the blue wing of the spectrum, while in the red wing CE intensifies. This paves a way to tailor supercontinuum angular structure for applications.

3. Enhanced pulse-to-pulse stability and robustness against the beam aberrations is demonstrated for multifilament array with a Dammann grating compared to one created with a 4-lobe phase plate [2].

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Experimental study of terahertz emission from single-color filament

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Plasma formed during filamentation of laser pulses is one of terahertz radiation sources [1]. The simplest scheme for terahertz generation is single-color filamentation in air. However, the mechanisms and properties of the terahertz emission from singlecolor filament have been poorly studied due to relatively low efficiency.

In our experiments we used laser pulses with 740 nm wavelength, 90 fs duration and energy up to 5 mJ. To create filament plasma focusing elements with different numerical apertures were used. We detected terahertz emission with a bolometer sensitive in the range of 0.1 - 12 THz. To distinguish various frequencies, we set narrowband terahertz filters in front of the bolometer input window. By moving the bolometer around the plasma channel, we measured two-dimensional angular distributions of terahertz radiation.

Figure 1. Normalized two-dimensional angular distributions of terahertz radiation at frequency of 0.3 THz (a), 1 THz (b) and 10 THz (c). The pulse energy is 3 mJ, the numerical aperture is 0.02.

For example, Fig. 1 shows normalized two-dimensional angular distributions of terahertz radiation at different frequencies. At a frequency of 0.3 THz (Fig. 1a), terahertz radiation propagates into a cone with a minimum on the axis. At a frequency of 1 THz (Fig. 1b), the pattern structure takes the form of two maxima located on the axis perpendicular to laser polarization. At higher frequencies the cone-shaped structure is restored again. Figure 1c shows the distribution at 10 THz.

It should be noted that the propagation angles of terahertz radiation at different frequencies differ significantly and for low frequencies can reach more than 30 degrees. In addition to information about the propagation structure, energy characteristics of terahertz radiation can be obtained from two-dimensional patterns by integrating signals over the distribution. This approach allows to take into account terahertz radiation propagating at wide angles. Thus, we investigated terahertz patterns, spectral and energy characteristics of terahertz emission under various laser parameters such as wavelength, pulse duration, numerical aperture and energy.

The work is supported by Russian Science Foundation grant #24-19-00461.

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Long distance laser clearing of fog for free space optical communications (FSO)

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Laser filamentation has recently emerged as a possible solution for creating optically clear channels in fog (DelaCruz2016, Schimmel2018). These demonstrations were performed on the laboratory scale with small cloud chambers. Nevertheless, during the field experiment on the Santis Mountain which demonstrated lightning guiding over 60 m with filaments (Houard2023), it was observed that laser lightning guiding was also effective in fog conditions, which pointed to long distance cloud clearing as well. To this end, we carried out a horizontal experiment at the Laboratoire de l'Accelerateur Lineaire (LAL) in Orsay using the same laser as the one that was implemented on the Santis (1030 nm, 500 mJ, 1 ps, 1 kHz). Two mobile cloud chambers of 3 m and 10 m length respectively could be moved along the laser path, with different cloud droplet concentrations to produce a wide range of attenuation.

Figure 1. Cloud clearing experiments with filaments optimized at 50m (left) and 100 m (right)

We found that the bundle of filaments could span over 40 m, when temporal and spatial focusing was optimized. The transmission through the fog chambers was assessed by direct laser power measurements and by the superposition of a cw laser transmitter, a photodiode and a camera. Figure 1 shows the results, with a spectacular 10 dB transmission gain, even with relatively modest energies (100 mJ). It was also observed that the heat deposited by the filament bundle induced significant lensing and turbulence on the cw laser transmission. The filament induced turbulence has been modelled with high level fluid dynamics computations and the agreement with the experiment is very satisfactory. It is planned to use the computational model to tailor spatio- temporal characteristics of the clearing laser in order to minimize turbulence or even guide the laser that will be used for FSO.

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Polarization dynamics of femtosecond laser pulse during filament formation

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The polarization of the femtosecond laser pulse plays an important role in laser filamentation, which influences the plasma formation, supercontinuum generation, lasing action, etc. It has been theoretically reported that the evolution of the pulse waveform along the propagation distance has a tight correlation with the initial polarization parameters. Therefore, measuring the polarization dynamics of the ultrashort laser pulse in the filamentation regime could provide useful guidance for certain polarization-sensitive processes, such as backward lasing enhancement and high-order harmonic generation. Here, we reported recent experimental and theoretical studies on the polarization dynamics of the filament-forming near-IR femtosecond laser pulse in a large parameter space of initial polarization, pulse energy, and gas pressure. It was observed that an initially circularly polarized pulse cannot sustain its polarization during filamentation. Theoretical simulations largely reproduce the observations.

Figure 1. Final polarization ellipticity versus initial rotation angles (0 for linear, 45 for circular) for different laser pulse energies under (a) 0.1 kPa and (b) 2.0 kPa.

Frequency resolved terahertz beam from femtosecond filament in air

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Femtosecond air-based plasma is known as a source of wideband terahertz (THz) radiation $[1]$. There are three commonly used air-based THz generation schemes: femtosecond single-color plasma channel (unbiased filament) $\left[1,2\right]$, the single-color plasma channel placed in the external electrostatic field (DC-biased filament)^[2] and the two-color plasma channel^[4,5]. The single – color THz generation schemes do not require ω – 2 ω phase adjustment and can be scaled up to terawatt peak power optical pump^[6]. Since in THz spectroscopy a certain area of the sample should be illuminated, the knowledge of the applied THz electrical field strength provided by THz beam shape on the sample is important^[7] in addition to the integrated THz yield.

In this work we measure and simulate 2*D* frequency-resolved THz far field beam profiles for a single-color^[8], single-color DC-biased^[9] and two-color^{[\[10\]](#page-49-5)} filament formed by geometrically focused 740-nm, 90-fs pulses. THz radiation was detected in two dimensions by superconducting bolometer. Simulations were performed with 3D axially symmetric unidirectional pulse propagation equation (UPPE)^[9,10] or far-field THz vectorial distribution model for a single-color filament^[8].

In a single-color unbiased filament THz beam distribution reveals strong dependence on filter bandpass frequency. Axially symmetric radially polarized distribution breaks up into two lobes in the vicinity of ∼1 THz or exhibits azimuthal modulation at the filter frequencies up to 10 THz.

THz far field beam distributions from the DC-biased filament are flat-top with FWHM of 10–30 degrees for the range 0.3-3 THz and conical at ∼10 THz. The flat-top beams are good candidates for THz spectroscopy applications requiring homogeneous illumination of the sample.

Figure 1. Angular distributions of 0.3-THz fluence for the phase φ corresponding to the maximal (a) and minimal (b) THz yield in the experiment (circles) and simulations (curves). Panels (a) and (b) are in the same scale

THz beam from the two-color filament demonstrates robust ring carrying 80% of the overall THz yield independently of the $\omega - 2\omega$ phase offset φ . Only the on-axis THz

radiation is sensitive to \mathcal{P} [Fig. 1] and can be neglected in the experiment for the sake of the more energetic ring.

The research is performed under the financial support of the Russian Science Foundation (24-19-00461).

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Conductivity measurement of warm-dense gold with single-shot terahertz time-domian spectroscopy

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Terahertz (THz) waves offer a distinctive diagnostic method for detecting high .energy density matter [1]. However, realizing the THz time-domain spectral (THz-TDS) diagnosis of matter states under extreme conditions in large high-energy density devices remains a significant obstacle. To address this requirement, we designed and implemented an optical pump-THz single-shot detection system driven by a strong femtosecond laser. In the experiment, the $LiNbO₃$ wafer with the diameter of 3 inches generated THz radiation with pulse energy of 1 milli-Joule and electric field of 10MV/cm. The strong THz pulse was employed to measure the transient THz conductivity of 30 nm thick freestanding gold foils pumped by a 400 nm laser pulse. The system possesses the capability of THz single-shot diagnosis of nonequilibrium states of matter under extreme conditions,which promotes the strong THz field phsics.

Figure 1. THz waveforms transmitted through the 30-nm-thick golden film (left) and the measured resistivity (right) at different pump delays.

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GW-TW terahertz radiation and its propagation in media

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High-power terahertz (THz) radiation sources are essential for many applications. Nevertheless, the generation of gigawatt-to-terawatt THz pulses remains thus far a formidable challenge. Ultraintense laser-produced plasmas have attracted everincreasing interest as a damage-free medium for generating high-peak-power THz pulses. In this talk, recent progresses on the ultraintense laserdriven THz radiation from plasmas will be reviewed [1]. TW-scale THz radiation has been demonstrated experimentally by irradiating a thin solid foil with a high-intensity laser pulse [2]. By tuning the laser or target parameters, the THz waveform and spectrum can be manipulated effectively [3], and the laser-to-THz energy conversion efficiency can be boosted up to \sim 1% [4]. The propagation of such extreme THz pulses in transparent media like the dispersionless gases and the dispersive dielectrics is investigated numerically. It is found that, the self-focusing of THz pulses in the weakly ionized gases could occur at a power much lower than the critical power for Kerr self-focusing, P_{cr} , while much higher power than P_{cr} is required for the self-focusing of fewcycle THz pulses in a non-ionized dielectric medium.

Figure 1. Overview of THz peak powers versus pulse energies for various intense pulsed THz sources [1].

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Ultrafast laser filamentation in semiconductors

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Ultrafast laser filamentation in fluids can advantageously be used for countless applications including light detection and ranging (LIDAR) [1], spectroscopy [2], terahertz wave generation [3], fog clearing [4], air waveguides [5], and lightning guidance [6]. However, in semiconductors exhibiting extreme nonlinear refractive indices, delocalization of the energy deposition as well as intensity clamping represent major challenges for precise in-volume processing applications [7, 8]. To date, most studies on ultrafast laser filamentation in these solids have been performed in silicon.

We examine ultrafast filamentation in different semiconductors with nonlinear propagation imaging, as illustrated in Fig. 1(a). From these measurements, we obtain in-volume three- dimensional (3D) fluence distributions for various laser conditions [Fig. 1(b)]. Combining these measurements with modeling, different parameters related to energy deposition, nonlinear refraction and absorption are extracted. The saturation of the energy deposition as a function of the input pulse energy [Fig. 1(c)] demonstrates that filamentation universally dictates propagation in all tested semiconductors. This new understanding paves the way to the future development to maximize energy deposition in semiconductors.

Figure 1. (a) Nonlinear propagation imaging. (b) Fluence distributions in different semiconductors for various input pulse energies. The radial and on-axis scale bars are 10 µm and 100 µm, respectively. (c) Evolution of the in-volume maximum fluence as a function of the input pulse energy for different semiconductors. The dotted lines are calculations in the linear propagation regime.

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High-resolution and high-speed molecular spectroscopy with optical combs

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Optical frequency comb that contains hundreds and thousands of evenly-spaced frequency elements (or comb lines) has advanced approaches to optical metrology, including molecular spectroscopy. We will report on our recent works on direct frequency comb spectroscopy, i.e. dual-comb spectroscopy (DCS). In fact, DCS is one of the most promising comb-enabled spectroscopic techniques for nondestructive interrogation of, e.g., molecular structures, with unprecedentedly high spectral resolution and high acquisition speed. With two frequency combs of slightly different line spacing beating on a single detector, it enables simultaneous detection of multiple molecular transition lines covered in the wide spectral range of the combs. Here, dualcomb spectroscopy with two electro-optic frequency combs tunable in the near-infrared and high-wavenumber Raman fingerprint region will be reported for molecular sensing

Fig. 1 Ultra-rapid mid-infrared molecular spectral results [1]

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Nonlinear post-compression in solid-state multipass cells

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Nonlinear post-compression plays an important role in generating high-energy, ultrashort pulse. One of the most powerful methods for nonlinear post-compression is using the multipass cells, which has attracted increasing attention since 2016. While many experimental work have demonstrated the nonlinear pulse compression in solidand gas-filled multipass cells. Some deep mechanisms about the process of nonlinear interaction need to be revealed. In this talk, we will show the theoretical aspects of nonlinear pulse compression in solid-state multipass cells. The coherence about the supercontinuum generation, the thermal effects as well as the mode-resolved nonlinear processes will be discussed.

Quantum optimal control of air lasing at ambient conditions

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Coherent generation and amplification of ultraviolet light, which occurs during filamentation of infrared laser pulses in ambient air, is known as air lasing [1]. When observed at atmospheric pressure, this phenomenon is, arguably, among the most spectacular and unexpected manifestations of quantum coherence and inversionless lasing. Indeed, while lasing without inversion typically requires careful arrangement of the parameters of several laser fields, air lasing at atmospheric pressures appears to arise naturally, without special efforts, and under a broad variety of laser filamentation conditions [2]. We present a lasing-without-inversion mechanism [3,4] that is active in the 391 nm amplification process in nitrogen molecular ions under standard conditions where femtosecond laser filamentation leads to self-guiding of light. We show that the lasing without inversion is triggered by the combination of strong-field molecular ionization and molecular alignment, i.e., it arises naturally during the propagation of intense femtosecond laser pulses in the air, using only the natural dynamics of a multilevel quantum system. However, the lack of population inversion in air lasing at ambient conditions also limits its potential. In addition, I will present experimental results [5] which show how a sequence of welltimed laser pulses can be used to enhance the air lasing efficiency by orders of magnitude. Our theoretical analysis [5] shows that an optimal pulse sequence generates a true population inversion in nitrogen molecular ions. Importantly, the approach enables population inversion and gain without requiring fine tuning of the pulse parameters. Thus, it represents a major step forward towards achieving robust backward UV lasing during filamentation of infrared pulses, which could finally enable application of IR-UV air lasing in remote sensing.

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Adding the third harmonic: effect on ionization, terahertz emission, and harmonic generation

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Ultrafast strong-field ionization is a basic phenomenon that intertwines with laser filamentation, generation of ultrabroadband terahertz pulses, and harmonic generation. One of the ways to modify or control the strong-field ionization and the associated ionization-induced phenomena is to employ two-color pulses containing second harmonic along with the pulse at the fundamental frequency. The relative phase of the second harmonic with respect to the fundamental field can be controlled in experiment by changing the optical path of the two-color pulse inside the dispersive medium. This allowed researchers to obtain terahertz pulses with unique parameters as well as to improve yield of high-order harmonics in the experiments. In recent years, several studies explored experimentally and theoretically the use of ionizing pulses containing also the third harmonic beside the fundamental and second ones.As it turned out, adding the third harmonic may be beneficial in many aspects: controlling ionization and filamentation^[1, 2], enhancing terahertz emission^[3, 4], and modifying the harmonic spectra^[5, 6].

This talk summarizes and generalizes the recent results of studying the effects induced by ultrafast three-color strong-field ionization of gases in context of controlling (enhancing and suppressing) ionization $^{[7]}$, broadband terahertz emission $^{[8, 9]}$, and the low-order harmonic spectra [10] It appeared that even very weak third harmonic (at the intensity level of 10^{-2} of the fundamental field and lower) may significantly alter the ionization probability and the spectral composition of the secondary emission.The presence of the third harmonic with controllable phase delay also significantly expands the capabilities for diagnostics of laser-created plasma and ionization dynamics as well as for shaping the spectra of the secondary radiation. These capabilities may be attributed to the competition between different parametric channels present due to the degenerate character of wave mixing processes. The ability to adjust the ionization probability in experiments also opens up ways to finely tune and optimize various phenomena accompanying laser-material interactions: filamentation and intensity clamping, high-order harmonic and attosecond generation, nanofabrication, and remote ablation of samples.

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Spatiotemporal solitons in air-plasma channel

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Intense ultrashort pulses with durations down to a few optical cycles have been strongly demanded by ultrafast science and strong-field physics. However, high-peakpower few-cycle pulses, especially sub-two-cycle pulses, are usually accompanied by one or several temporal sidelobes, which seriously restrict their applications. Despite the quality improving approaches including additional third-order dispersion compensation^{[\[1\]](#page-61-0)} and the pressure gradient^[1], their parameter sensitivity makes it still a challenge to obtain intense few-cycle pulses with a high temporal quality.

In this work, we propose a unique scheme to improve the temporal quality of fewcycle pulses by the self-cleaning of spatiotemporal solitons in a preformed air-plasma $channel^[3]$.

Figure 1. Normalized spatiotemporal intensity evolution for the 1.5 μm laser pulse in a preformed airplasma channel.

Based on the negative dispersion introduced by the preformed plasma channel, a long-distance spatiotemporal soliton is formed after the coalescence of the two subpulses, as shown in Fig. 1. At the initial stage of the soliton regime, there are sidelobes both before and after the main pulse. During the long-distance propagation, the highintensity soliton ionizes the air. The resulting blue shift causes the velocity of soliton to continuously increase. Consequently, the front sidelobes are captured by the soliton. Moreover, the plasma generated by the soliton continuously defocuses the ubsequent sidelobes into the background. Therefore, the sidelobes are eliminated, and the temporal profile of the few-cycle pulse is self-cleaned after a long-distance propagation. By changing the density of the preformed air-plasma channel, this scheme can be applied to compress femtosecond laser pulses with different central wavelengths to sub-twocycle pulses with a high temporal quality.

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Generation of Spatiotemporal Optical Vortex Attosecond Pulse Trains

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The realization of spatiotemporal vortex structure of various physical fields with transverse orbital angular momenta (OAM) has attracted much attention and is expected to expand the research scope and open new opportunities in their respective fields. Here we present theoretically the first study on generation of attosecond pulse trains featuring a spatiotemporal optical vortex (STOV) structure by a two-color femtosecond light field, with each color carrying transverse OAM. Through careful optimization of relative phase and intensity ratio, we validate the efficient upconversion of the infrared pulse into its tens of order harmonics, showing that each harmonic preserves a corresponding intact topological charge. This unique characteristic enables the synthesis of an extreme ultraviolet attosecond pulse train with transverse OAM. In addition, we reveal that ionization depletion plays an outsize role therein. Our studies pave the way for generation and utilization of light fields with STOV in the attosecond regime.

Yield (arb. units) $|E_{\text{real}}(X,t)|^2$ (arb. units) angle $[E_{\text{real}}(X,t)] - \omega_{\text{real}}t$

Figure 1. Left column: spatially-resolved HHG spectra driven by STOV pulses. Middle column: retrieved spatiotemporal intensity distributions. Right column: retrieved spatiotemporal phase distributions. (a) Single-color STOV driving field; (b) Two-color STOV driving field with unoptimized phase; (c) Two-color STOV driving field with optimized phase.

Figure 2. The retrieved spatiotemporal intensity distributions of attosecond pulse trains: (a) Single-color STOV driving field; (b) Two-color STOV driving field with unoptimized phase; (c) Two-color STOV driving field with optimized phase; (d) The temporal intensity profile of a single attosecond pulse sampled from the black rectangle in (c).

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