

New Horizons of Optics of the Midinfrared Spectral Range

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Abstract—Optical physics and laser technologies are rapidly moving in the direction of exploring the midinfrared spectral range. New methods of mid-IR ultrashort pulse generation allow forming very short flashes of electromagnetic radiation with record high peak power for this range. The first experiments conducted with such systems make possible implementing new regimes of laser–matter interaction and shed light on unusual properties of the nonlinear-optical response of materials in the mid-IR spectral range.

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INTRODUCTION

Advancing into the midinfrared (mid-IR) spectral range is one of the most challenging tasks of laser technologies and optics of ultrashort laser pulses. There is considerable interest in this range [1, 2] because of the possibilities of using powerful ultrashort pulses of low-frequency radiation for substantial increase of efficiency of laser acceleration of particles and generation of optical harmonics; increasing thresholds of unwanted nonlinear-optical processes; and implementing new methods of spectroscopy and microscopy of physical, chemical, and biological objects. Despite the strong motivation for investigating the mid-IR spectral range, optical experiments focusing on studying ultrafast processes in matter on the femtosecond time scale have been limited to visible and near-IR spectral ranges, in which effective laser sources of ultrashort pulses were developed and successfully used.

The complexity of advancing laser technologies to the mid-IR spectral range is largely due to the fact that there are no efficient laser materials with gain bandwidth sufficient for femtosecond pulse generation in the mid-IR range, in contrast to near-IR range where laser sources of high-power ultrashort pulses were developed and successfully used in experimental studies. However, research carried out in the past 2–3 years has demonstrated that the problem of developing sources of ultrashort mid-IR pulses can be suc-

cessfully solved by using nonlinear-optical parametric frequency conversion of ultrashort pulses. In particular, the technology of optical parametric chirped-pulse amplification was recently used for generation of ultrashort laser pulses with wavelength of about 4 μm [3] and energy approaching 10 mJ. The first experiments on studying higher-order nonlinearities in the atmosphere and inert gases were conducted with the help of laser sources of this class [4, 5]. The possibility of higher-order harmonics generation that allows obtaining coherent X-ray emission with photon energies of up to 1.3 keV was also demonstrated [6]. The peak power of mid-IR ultrashort pulses, which can be obtained with sources of this class, allows achieving filamentation of a beam of electromagnetic radiation in high-pressure gases [5, 7] and observing the onset of laser oscillation in nitrogen molecules under such conditions [8]. The data obtained in these experiments and supported by theoretical studies [9–11] suggest the existence of new regimes of laser–matter interaction in the mid-IR spectral range.

In the present work, we briefly review the first experiments conducted with high-power ultrashort mid-IR pulses, generation of which became possible due to advent of a new generation of laser systems based on the technology of optical parametric chirped-pulse amplification. Experimental studies conducted with femtosecond pulses of this class demonstrate the possibility of transmission of pulses of electromagnetic radiation with energy exceeding

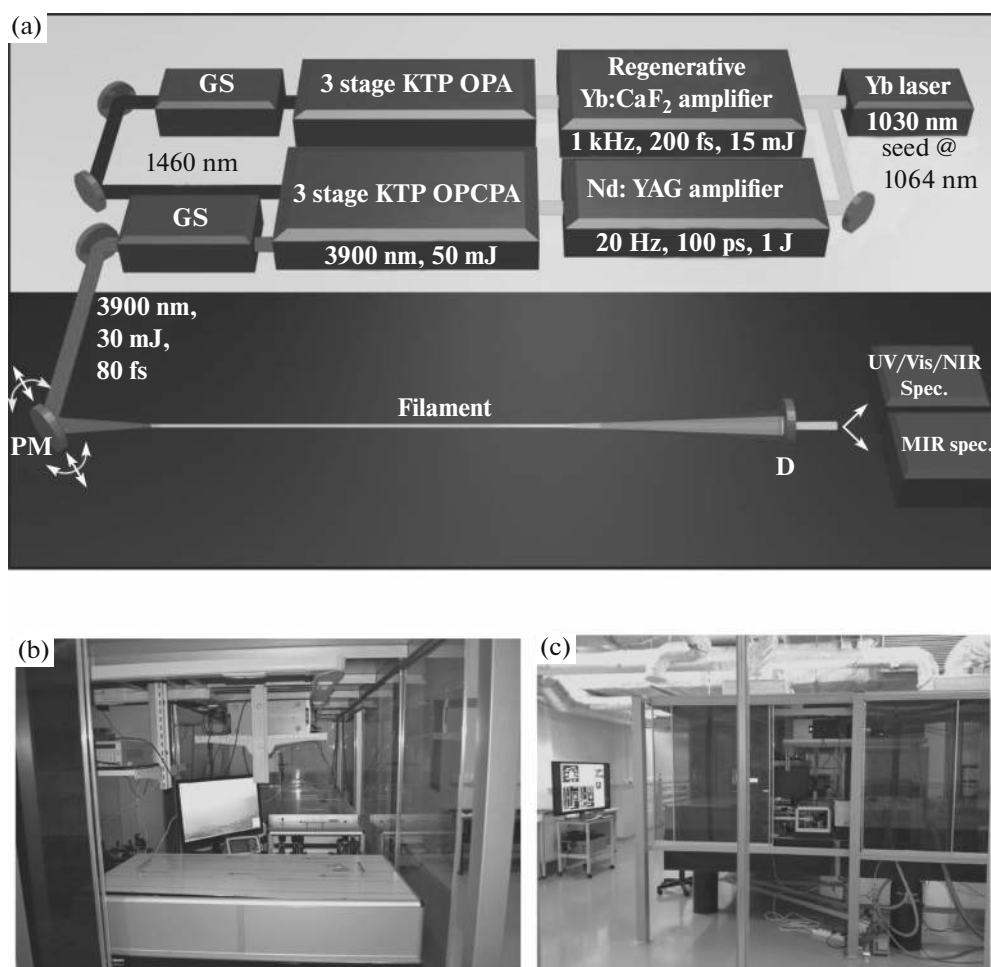


Fig. 1. (a) Layout and (b, c) photographs of the source of high-power ultrashort mid-IR pulses. The photographs were taken in the Laboratory of Photonics of the Russian Quantum Center.

20 mJ through the atmosphere in the single-filament regime. New regimes of optical harmonics generations were implemented in the presented experiments. The conditions of compression of subterawatt mid-IR pulses down to a few cycles of the light field through filamentation are investigated, and methods of its implementation are proposed. The possibilities of remote sensing of the atmosphere by using the new physical phenomenon of laser oscillation in filaments created by mid-IR ultrashort laser pulses are discussed.

SUBTERAWATT SOURCE OF MID-IR FEMTOSECOND PULSES

In this work, we used a laser system comprised of a solid-state ytterbium oscillator, a regenerative amplifier, a three-stage optical parametric amplifier, and a three-stage optical parametric chirped-pulse amplifier for obtaining high-power ultrashort pulses of electromagnetic radiation in the mid-IR spectral range (Fig. 1). An Yb:CaF₂ oscillator generated ultrashort pulses with a spectrum centered at 1030 nm [12].

Regenerative amplification of these pulses in our system could produce pulses with energy of up to 15 mJ with a repetition rate of 1 kHz and pulse duration of less than 200 fs. Pulses of radiation with a central wavelength of 1030 nm, pulse duration of less than 200 fs, and energy that could be varied from 3 to 12 mJ were used in the experiments presented below for studying filaments in the near-IR spectral range.

Regeneratively amplified pulses with a central wavelength of 1030 nm, pulse energy of about 1 mJ and duration of about 190 fs were used for generation of ultrashort pulses in the mid-IR spectral range in the scheme of a three-stage parametric amplification. The pulses at the output of the optical parametric amplifier had central wavelength of 1460 nm and duration of about 200 fs. After stretching in a prism stretcher, the pulses were used as a signal wave in a three-stage optical parametric chirped-pulse amplifier comprising three successive KTA crystals optically pumped by pulses of a Nd:YAG laser with duration of 100 ps. The pump laser output was split into three beams delivering pump energies of 50, 250, and 700 mJ. The energy of the idler wave at the output of the final stage of the

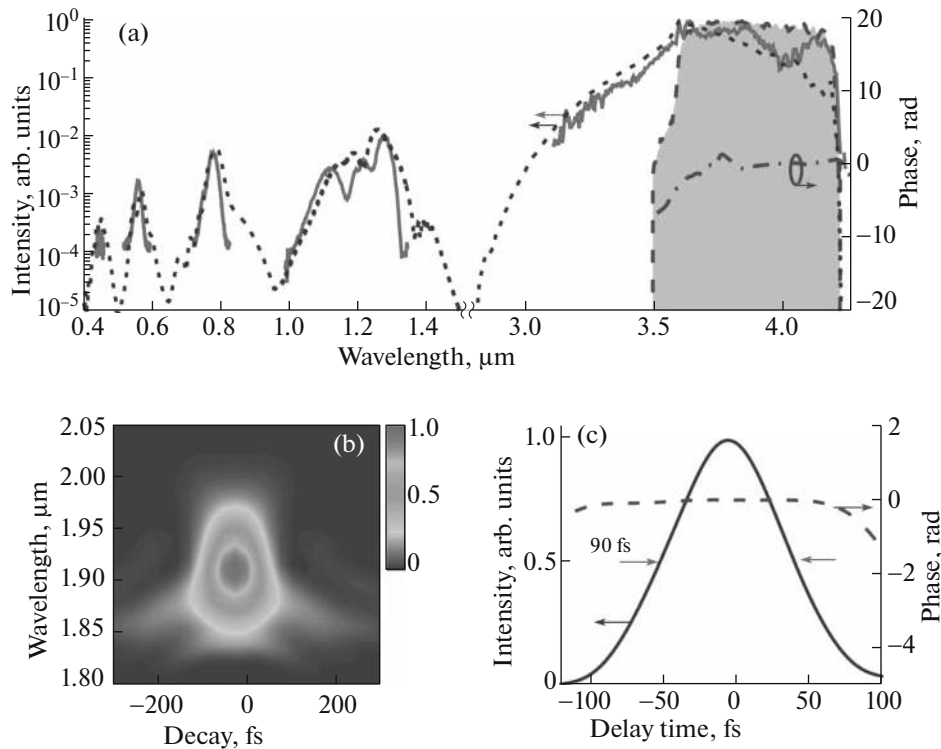


Fig. 2. Spectrum (dashed line with filling) and spectral phase (dash-dotted line) of the idler wave formed at the output of the optical parametric chirped-pulse amplifier, along with the experimental (solid line) and simulated (dashed line) spectra of radiation emerging from (a) the filament formed in the atmosphere by pulses of radiation with central wavelength of 3.9 μm, duration of 90 fs, and energy of 22 mJ after focusing by a lens with focal distance of 75 cm. (b) A FROG spectrogram and (c) temporal pulse envelope and phase of the idler wave retrieved from it formed at the output of the optical parametric chirped-pulse amplifier.

optical parametric chirped-pulse amplifier exceeded 50 mJ [13, 14]. After pulse compression in a grating compressor, we obtained pulses with a central wavelength of 3.9 μm, energy of up to 30 mJ, and duration between 80 and 200 fs.

Spectral measurements in the mid-IR spectral range were performed by means of a scanning monochromator and a thermoelectrically cooled IR detector based on HgCdTe. Standard Ocean Optics spectrometers were used for spectral measurements in the visible and near-IR spectral range. Temporal characterization of output pulses with phase retrieval was implemented by means of the second-harmonic generation frequency resolved optical gating (FROG), wherein second harmonic was generated in a 0.5-mm-thick AgGaS₂ crystal. Characteristic power spectrum of an ultrashort pulse of the idler wave obtained at the output of the optical parametric chirped-pulse amplifier is shown by a dashed line with filling in Fig. 2a. A characteristic FROG spectrogram of the pulse is presented in Fig. 2b. The time envelope and phase of the pulse retrieved from this FROG spectrogram are shown in Fig. 2c.

LASER FILAMENTATION IN THE MID-IR SPECTRAL RANGE

The implemented technology of optical parametric amplification allows achieving qualitatively new peak power level of mid-IR femtosecond pulses. Peak powers of mid-IR femtosecond pulses sufficient for forming laser filaments under normal atmospheric conditions were obtained for the first time in the experiments presented in [13]. To generate a laser filament in the medium exhibiting optical nonlinearity coefficient n_2 and refractive index n_0 , the peak power of the ultrashort laser pulse must be considerably higher than the critical power for self-focusing, $P_{cr} = C(8\pi n_0 n_2)^{-1} \lambda^2$, where λ is the wavelength and C is a numerical factor ($3.72 < C < 6.4$) determined by transverse intensity profile of the laser beam. In the experiments conducted previously, filamentation of ultrashort mid-IR laser pulses was observed in high-pressure (at the level of several atmospheres) gases. The peak power of previously available laser systems was insufficient for generation of mid-IR laser filaments under atmospheric conditions.

The regime of filamentation of mid-IR laser pulses under atmospheric conditions was achieved for the first time in [13]. The experiments demonstrated the

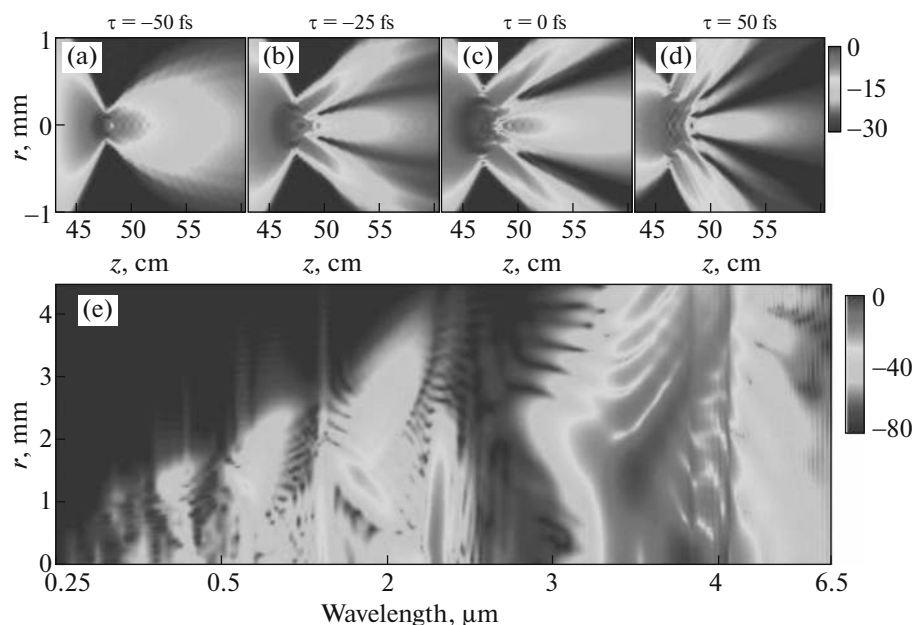


Fig. 3. Spatiotemporal dynamics of intensity of subterawatt ultrashort mid-IR pulse in the atmosphere at the (a, b) pulse front, (c) center, and (d) tail. Solid line shows beam diameter determined at half-maximum of its intensity in the beam cross section. (e) Spectral distribution of supercontinuum across the beam.

possibility of generating laser filaments in the atmosphere by ultrashort pulses of electromagnetic radiation with a central wavelength of 3.9 μm , duration of 80–130 fs, and peak power between 0.2 and 0.3 TW [14]. When pulsed mid-IR laser radiation with such parameters was loosely focused in the atmosphere by a set of lenses fabricated from calcium fluoride, a bright extended spark stemming from effective ionization of atmospheric air was observed, along with substantial broadening of the IR pulse spectrum (Fig. 2a).

The effect of filamentation in the atmosphere was observed in our experiments in a wide range of focal distances of the lens focusing mid-IR radiation. For given peak power and pulse duration, the length of the filament, peak intensity of electromagnetic field in the filament, and electron density in the filament are determined by the focusing conditions. Numerical simulations based on the model of filamentation of ultrashort pulses of mid-IR spectral range [5, 7, 13] showed that peak electron density drops by more than an order of magnitude upon increasing focal length of the lens focusing IR radiation from 45 to 200 cm. In so doing, the length of the ionized area increases from several centimeters to 1.5 m.

SPECTRAL EVOLUTION OF SUBTERAWATT MID-IR PULSES: STRONG SUPERCONTINUUM IN THE MID-IR SPECTRAL RANGE AND OPTICAL HARMONICS

Since filamentation of ultrashort mid-IR pulses is accompanied by many new effects not seen in the case

of laser filaments in the near-IR spectral range, it would be advantageous to compare the physical picture of filamentation in mid-IR spectral range with a well-studied scenario of filamentation in the near-IR spectral range [15, 16]. Similar to the case of filamentation in the mid-IR spectral range [15, 16], the filaments formed by ultrashort pulses of the near-IR spectral range are characterized by effective spectral broadening caused by nonlinear-optical interactions that are strongly enhanced by conditions existing in the laser filaments due to suppression of diffraction-limited divergence of the laser beam over an extended optical length. However, spectral broadening in the long-wavelength part of the spectrum for pulses with a central wavelength of 3.9 μm is limited by the molecular absorption bands of the atmosphere. As a result, intense red wing of the spectrum, which is well pronounced in the case of filaments in the near-IR spectral range, becoming stronger with increasing laser pulse energy, turns out to be strongly suppressed in the case of filaments of the mid-IR spectral range.

The situation in the high-frequency part of the spectrum is opposite. The high-frequency wing of the spectrum of radiation emerging from the mid-IR laser filaments turns out to be strongly enhanced due to generation of optical harmonics. Favorable conditions for optical harmonics generation of the mid-IR pump field are explained by weak dispersion of the atmospheric air in the mid-IR spectral range. As a result, phase mismatch for generation of a group of several low-order optical harmonics in the case of mid-IR pump field is much smaller than analogous phase mismatch for generation of optical harmonics of the near-

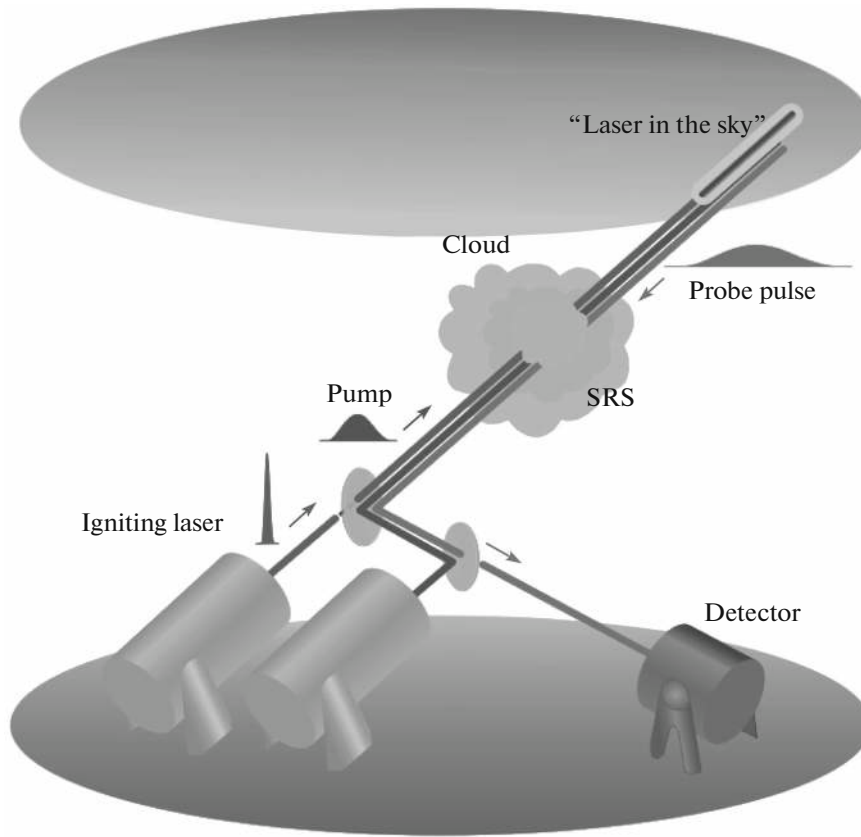


Fig. 4. Remote sensing of the atmosphere based on laser action in the filament.

IR pump field. An important additional factor facilitating generation of optical harmonics of the mid-IR pump field is the fact that a group of harmonics (including the third, the fifth, the seventh, and the ninth harmonics) of the mid-IR pump field falls into the transparency window of the atmosphere.

Theoretical analysis shows that the phenomenon of filamentation leads to enhanced generation of harmonics of mid-IR radiation due to suppression of diffraction-limited divergence of the pump beam, which results in extension of the area of effective nonlinear interaction of the pump and optical harmonics waves, in which high efficiency of the pump field is preserved. Spectra obtained at the output of the filaments generated by high-power ultrashort mid-IR pulses reveals intense peaks (Fig. 2a) corresponding harmonics of the pump field, which are broadened due to cross-phase modulation and shifted as a result of propagation through rapidly ionizing gas.

SPATIOTEMPORAL DYNAMICS OF SUBTERAWATT ULTRASHORT MID-IR PULSES IN THE ATMOSPHERE

For better understanding of filamentation of ultrashort mid-IR pulses, we plotted maps of the field

intensity distribution in the filament at the pulse front (Figs. 3a, 3b), its central part (Fig. 3c), and tail (Fig. 3d). The results show that the beam dynamics is different for different parts of the pulse. This difference is related to the dynamics of electron density that increases from the pulse front to its end. The pulse front causes ionization of air, forming transverse profile of electron density that drops from the beam center toward its periphery. Such an electron density profile leads to defocusing of the central part of the beam and, in particular, its tail. These phenomena are clearly seen in the maps of field intensity depicted in Fig. 3. For sufficiently long filaments, scattering of IR radiation from plasma formed by the field, which becomes particularly strong at the pulse tail, causes considerable pump depletion along the filament.

Beam diameter d defined as the full width of the beam cross section at half its maximum intensity is marked by a solid line in Figs. 3a–3d. Variation of beam diameter d along the path of the IR pulse allows defining the length of the experimentally generated filament as the distance between the points along the beam path in which the beam diameter is equal to twice the minimum beam diameter d . In the case of overly tight beam focusing, strong scattering of the central (Fig. 3c) and tail (Fig. 3d) sections of the pulse

from the electron density profile created by the pulse front limits of the length of the filament occurs. Proper choice of focusing conditions (Figs. 3a–3d) allows balancing beam self-focusing and defocusing caused by transverse electron density distribution. It can be seen from Figs. 3a–3d that, in this regime, the length of the filament can reach several meters, which opens unique possibilities for remote sensing of the atmosphere and long-distance transmission of high-power laser pulses.

LASER ACTION IN A FILAMENT: NEW POSSIBILITIES OF REMOTE SENSING OF THE ATMOSPHERE

The most successful and widely used methods of remote sensing are based on using incoherent light scattering (Rayleigh scattering, Mie scattering, or Raman scattering) and emission of incoherent optical radiation [17, 18]. The intensity of incoherent optical signal rapidly decays with distance, which imposes considerable limitations on sensitivity of the optical remote sensing methods. These problems can be radically solved by using coherent optical signals for remote sensing [19]. The main difficulty blocking realization of this idea is related to fundamental limitations on generation of coherent optical signals in backward direction [19], which is a necessary condition for implementing remote sensing and signal detection by a remote sensor located next to a source of optical radiation (Fig. 4). For a broad class of coherent optical processes that are promising for solution of problems of remote sensing, such as four-wave mixing, coherent anti-Stokes Raman scattering, etc., this limitation reveals itself in the form of a ban on signal generation in the direction opposite to the direction of propagation of the pump fields, which is related to the requirement of conservation of momentum in optical interactions [19].

A number of promising schemes based on combination of laser and radar radiation [20–23], as well as using superradiance in the atmosphere [24], have been proposed to solve the problem of coherent signal generation in backward direction. An important step on the way to development of a remote source of coherent backward signal was the discovery of the effect of laser oscillation in atoms of atmospheric oxygen [25, 26]. The atmospheric oxygen laser can provide a strong backward optical signal [26] that can be used for solving the problems of remote sensing.

Currently, the key unsolved fundamental problem is implementation of laser oscillation under remote sensing conditions. In the experiments on laser oscillation in atoms of atmospheric oxygen, the laser effect could be initiated only at a distance of several meters [26]. Experimental and theoretical studies conducted in [8] allowed solving the problem of remote initiation of laser action in the atmosphere and controlling this process by using the effect of filamentation of ultra-

short mid-IR laser pulses. The authors experimentally demonstrated the possibility of remote chemically selective sensing of the atmosphere based on stimulated Raman gain or loss spectroscopy by using laser action in the filament.

The phenomenon of lasing in the filament demonstrated in experiments [8] opens unique possibilities for remote sensing of the atmosphere (Fig. 4). To demonstrate these possibilities, an experiment was conducted in which laser radiation generated in the filament was used as a probe field for detection of small impurities in gas mixtures by means of stimulated Raman gain or loss spectroscopy in counterpropagating beams (Fig. 4). Using stimulated Raman scattering (SRS) allows overcoming fundamental difficulties related to the requirement of fulfilling the phase-matching condition. The latter imposes fundamental limitations on applicability of a broad class of nonlinear optical phenomena, including coherent anti-Stokes Raman scattering, for schemes of remote sensing including counterpropagating beams [27]. Radiation of an auxiliary laser source propagating in the direction of the laser filament was used as a pump field in the implemented scheme of stimulated Raman gain and loss spectroscopy (Fig. 4). The obtained results open a new direction in the area of remote sensing of the atmosphere.

FIBER SYNTHESIZERS OF ULTRASHORT MID-IR PULSES

Pulse compression in hollow optical waveguides is one of the key technologies of modern optics of ultrashort pulses. This technology continued to develop in recent years, allowing one to obtain progressively shorter pulses with ever-increasing power. The combination of this technology with technologies of fabrication of microstructured optical waveguides [28, 29] opens up possibilities of obtaining mid-IR pulses with a duration of about one cycle of the field with peak power in the gigawatt range [30, 31]. The physical basis for generation of pulses exhibiting such unique set of parameters is provided by the unique dynamics of ultrashort optical breathing solitons. Understanding this dynamics requires going beyond standard approximations of the models based on solving the nonlinear Schrödinger equation (NSE).

It was demonstrated in [11] that the dynamics of ultrashort flashes of electromagnetic radiation with a duration of about one cycle of the field may be substantially different from the dynamics of NSE solitons. Optical shock waves generated by such extremely short waveforms of the field lead to effective generation of high-frequency components in its spectrum, which can be used for seeding the process of optical parametric amplification, provided that waveguide dispersion is chosen properly. This phenomenon leads to substantial spectral broadening of the formed pulse, thus

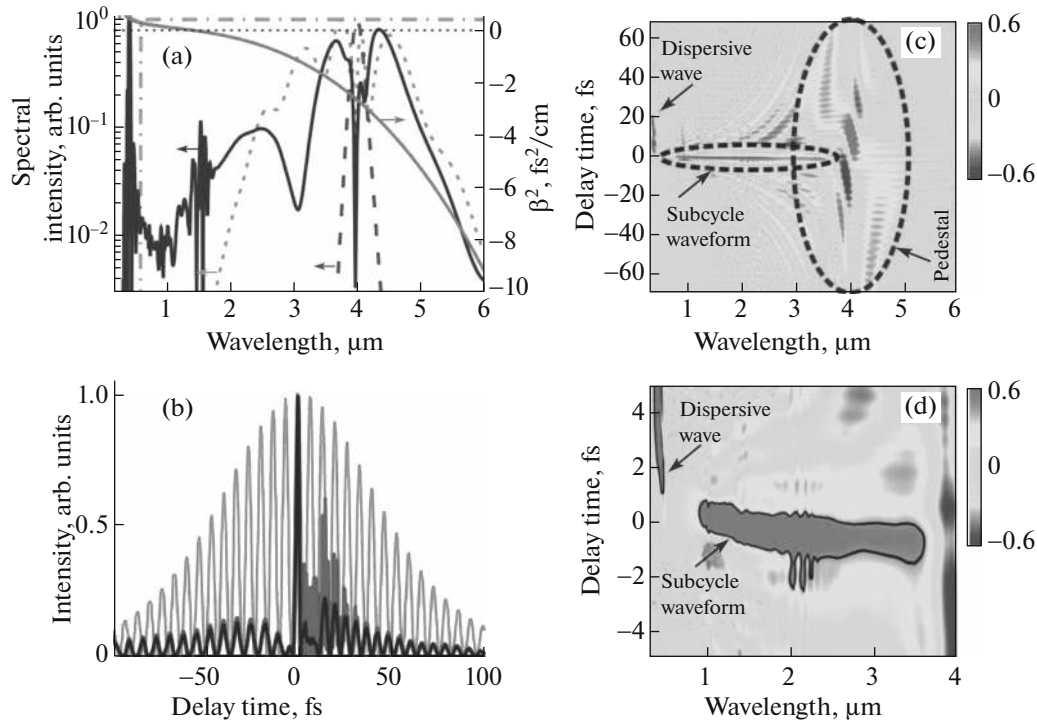


Fig. 5. (a) Spectrum of the pulse at the input of the waveguide (dashed line) and at the point of maximum compression, taking into account the presence of an optical shock wave (solid line) and the absence of one (dotted line). Also shown are the profile of waveguide dispersion and the spectrum of transmission of the optical filter used for selecting a pulse with a duration shorter than one cycle of the field. (b) Field intensity in the pulse before and after compression with and without spectral filtration. (c, d) Wigner spectrograms of compressed pulse at the point of maximum compression on (c) crude and (d) fine time scales.

allowing obtaining ultrashort pulses with a duration of less than one cycle of the field (Fig. 5).

Soliton self-compression of high-power mid-IR pulses in a hollow waveguide is illustrated in Fig. 5. Pulse compression is implemented in a hollow waveguide with an internal diameter of $500 \mu\text{m}$ filled by krypton at a pressure of 0.5 bar. The pulse at the input of the waveguide had a central wavelength of $4 \mu\text{m}$, duration of 100 fs, and energy of 2 mJ. The effects of higher-order dispersion caused the decay of the breathing soliton and the onset of soliton instability as a result of energy exchange between the soliton and the nonsoliton field component [32, 33]. To obtain very short pulses, the nonsoliton portion of field appearing in the form of an intense high-frequency peak in Figs. 5a and 5c is filtered out by a filter or due to specially selected transmission spectrum of the waveguide. Formation of the shock wave leads to substantial spectral broadening of the radiation (Fig. 5a), which allows forming pulses with duration of less than one cycle of the field (Figs. 5c, 5d). It was demonstrated in [34] that using hollow optical waveguides with a special kagome-lattice cladding allows implementing such a scheme of self-compression at the gigawatt level of peak power.

CONCLUSIONS

Optics of ultrashort laser pulses is rapidly advancing into the IR spectral range, opening new opportunities for implementing unique regimes of interaction of high-power coherent radiation with matter and generation of record-short pulses with duration in the atto- and zeptosecond range. Based on the technology of optical parametric chirped-pulse amplification in the field of picosecond laser pump pulses with energy at 1 J level, we generated mid-IR pulses with duration of less than 100 fs and peak power exceeding 0.3 TW. Experimental studies conducted with femtosecond pulses of this class demonstrate the possibility of transmitting pulses of electromagnetic radiation with energy over 20 mJ through the atmosphere in the single-filament regime. In the presented experiments, we implemented new regimes of optical harmonic generation, proposed methods of compression of subterawatt mid-IR pulses down to several periods of the light field through filamentation, and found the conditions for their implementation. We demonstrated the possibility of soliton self-compression of high-power femtosecond mid-IR pulses in hollow waveguides with a special cladding structure, which allows obtaining mid-IR pulses of gigawatt peak power with a duration close to one cycle of the electromagnetic field.

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