Nonlinear optics in the mid-infrared: new morning

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Abstract. Recent breakthroughs in ultrafast photonics in the mid-IR help understand complex interactions of high-intensity mid-IR field waveforms with matter, offer new approaches for x-ray generation, enable mid-IR laser filamentation in the atmosphere, facilitate lasing in filaments, give rise to unique regimes of laser–matter interactions, and reveal unexpected properties of materials in the mid-IR range.

1. Introduction
Motivated and driven by numerous applications and long-standing challenges in strong-field physics [1, 2], molecular spectroscopy, semiconductor electronics, and standoff detection, ultrafast optical science is rapidly expanding toward longer wavelengths [3-16]. Experiments reveal unique properties of filaments induced by ultrashort laser pulses in the mid-infrared, where the generation of powerful supercontinuum radiation is accompanied by unusual scenarios of optical harmonic generation, giving rise to remarkably broad radiation spectra, stretching from the visible to the mid-infrared. Generation of few- and even single-cycle mid-infrared field waveforms has been demonstrated within a broad range of peak powers and central wavelengths. Below-the-bandgap high-order harmonics generated by ultrashort mid-infrared laser pulses are shown to be ideally suited to probe the nonlinearities of electron bands, enabling an all-optical mapping of the electron band structure in bulk solids.

2. Generation of ultrashort pulses in the mid-infrared
High-power ultrashort mid-IR driver pulses are delivered in our experiments by a laser system (figure 1) consisting of a solid-state ytterbium laser with an amplifier, a three-stage optical parametric amplifier (OPA), a grating–prism (grism) stretcher, a Nd:YAG pump laser, a three-stage OPCPA system, and a grating compressor for mid-IR pulses [6, 8]. The compressed-pulse idler-wave OPCPA output has a central wavelength of 3.9 µm, a pulse width of 80 fs, and an energy up to 30 mJ.
measurements in the mid-IR range are performed with a homebuilt spectrometer consisting of a scanning monochromator and a thermoelectrically cooled HgCdTe detector. For the spectral measurements in the ultraviolet, visible, and near-IR ranges, OceanOptics HR4000 and NIRQuest spectrometers were employed. Temporal envelopes and phases of mid-IR pulses are characterized using frequency-resolved optical gating (FROG) based on second-harmonic generation (SHG) in a 0.5-mm-thick AgGaS$_2$ crystal.

![Laser facility with an OPCPA source of subterawatt ultrashort pulses in the mid-infrared at the Advanced Photonics Laboratory of the Russian Quantum Center, (b) ultrafast spectrochronography beamline, and (c) vacuum chamber for high-order harmonic generation.](image)

### Figure 1.

3. Laser filaments in the mid-infrared

Laser filamentation in the mid-infrared involves a broad class of nonlinear-optical phenomena that do not show up in visible and near-infrared filaments. The most prominent examples include generation of multiple odd-order harmonics [8] and strong plasma refraction [6], which tends to give rise to ring-shaped beam patterns, especially well-resolved toward the back of the pulse, as well as a shift in the balance between the self-focusing due to the Kerr effect and plasma-induced defocusing. With all these effects manifested in the inherently off-axial beam dynamics [6, 12], angle-resolved studies on the supercontinuum filament output have to be included as an essential tool needed to understand laser filaments in the mid-infrared. Since such angle-resolved measurements have to be performed within a bandwidth of several octaves, stretching from the ultraviolet deep into the mid-infrared, multiple challenges need to be confronted in implementing experimental methods that would enable such studies.
Our experimental approach addresses these challenges through angle-resolved studies on a multioctave high-energy supercontinuum output of laser filaments induced in atmospheric air by 0.25-TW, 3.9-μm driver pulses. With optical harmonics up to the 15th order contributing to supercontinuum generation alongside Kerr-type and ionization-induced nonlinearities, the spectra of this supercontinuum radiation span over almost five octaves from the ultraviolet deep into the mid-infrared spectral range. As we demonstrate below, rewards for such angle-resolved measurements are numerous. Performed on an ultrabroadband beam, these studies reveal clearly resolved signatures of a complex off-axial beam dynamics, which is intrinsic to laser filamentation in the mid-infrared, as opposed to near-infrared filaments, thus offering important insights into an intricate and in many ways unusual spatiotemporal evolution of high-power mid-infrared pulses in the filamentation regime.

Our experiments demonstrate the generation of high-energy supercontinuum spanning 4.7 octaves, from 250 to 6500 nm, using a 0.25-TW, 3.9-μm output of a mid-infrared optical parametric chirped-pulse amplifier as a driver inducing a laser filament in the air. The high-frequency wing of the supercontinuum spectrum is enhanced by odd-order optical harmonics of the mid-infrared driver. Optical harmonics up to the 15th order are observed in supercontinuum spectra as overlapping, yet well-resolved peaks broadened, as verified by numerical modeling, due to spatially nonuniform ionization-induced blue shift.

Filamentation-assisted pulse compression in the gas phase is shown to enable the generation of subterawatt few-cycle pulses in the mid-infrared. With both spatial modulation instabilities and excessive plasma scattering of the mid-infrared beam prevented through a careful choice of the gas pressure and the input peak power, providing single-filament regime of pulse propagation, peak powers as high as 0.3 TW are achieved in a truly single-mode, almost diffraction-limited 35-fs output at a central wavelength of 4 μm.

4. Ultrashort pulses in the mid-infrared and new regimes of nonlinear optics

Because of the complexity of the spatiotemporal field waveform dynamics underlying filamentation, experiments designed to guide theoretical modeling toward a detailed physical understanding of filaments generate high-volume, high-variety data sets at high data-acquisition speeds. Ultrabroadband, supercontinuum radiation spectra have to be measured in a filamentation experiment simultaneously with extremely complex beam patterns, pulse shapes, and field phase profiles. For a meaningful interpretation of an experiment, all these measurements have to be done online in a multidimensional parameter space, which involves varying the input peak power, pulse width, pulse chirp, beam diameter, beam shape, focusing geometry, as well as changing both linear and nonlinear material response within a continuum of dispersion profiles, nonlinear susceptibilities, ionization potentials, band gaps, etc. Since the spectra of supercontinuum radiation generated in laser filaments often span over several octaves, spectral characterization of such waveforms cannot be performed using a single spectrometer, requiring a set of accurately cross-calibrated spectrometers enabling spectral measurements in partially overlapping frequency ranges. Pulse characterization of such supercontinua cannot be done by standard methods, calling for a combination of innovative approaches. Beam characterization has to combine measurements of transverse intensity profiles with radially resolved spectral measurements, often augmented with fast-camera recording. To make this already very intricate picture complete, optical nonlinearities in filaments tend to amplify noise in both space and time, giving rise to beam and pulse breakup due to spatial and temporal modulations instabilities. As a result, multiple uncontrolled factors, such as random field intensity fluctuations across the beam and along the pulse, are a part of the picture. Processing this diversified information with a need to include an enormous number of both controlled and uncontrolled physical factors inevitably requires big-data management solutions.

Due to the computation complexity of numerical analysis of filamentation, especially at the level of peak powers orders of magnitude higher than the self-focusing threshold, where the assumption of axial symmetry of a beam fails and the full (3 + 1)-dimensional problem has to be solved, some of the
most interesting questions in the filamentation of high-intensity, high-peak-power ultrashort laser pulses open. Those include possible filamentation scenarios whereby high-intensity, high-peak-power ultrashort laser pulses available from the cutting-edge laser systems within a broad spectral range could be delivered over large distances, compressed in time, and coupled into regular or irregular arrays of quasistationary, quasisolitary spatiotemporal waveforms (Figure 2), including arrays of light bullets.

![Figure 2. Envisaged remote sensing scheme using high-peak-power solitons in the mid- and long-wavelength infrared: SPM, self-phase modulation; S, spectrometer.](image)

5. Conclusion
To summarize, our studies reveal unique properties of mid-infrared filaments, where the generation of powerful mid-infrared supercontinuum is accompanied by unusual scenarios of optical harmonic generation, giving rise to remarkably broad radiation spectra, stretching from the visible to the mid-infrared. Filamentation-assisted pulse compression in the gas phase is shown to enable the generation of subterawatt few-cycle pulses in the mid-infrared. Generation of few- and even single-cycle mid-infrared field waveforms with peak powers ranging from a few megawatts to hundreds of gigawatts has been demonstrated within a broad range of central wavelengths.

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