Phase-stable sub-cycle mid-infrared conical emission from filamentation in gases

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Abstract: Sub-single-cycle pulses in the mid-infrared (MIR) region were generated through a conical emission from a laser-induced filament. Fundamental and second-harmonic pulses of 25-fs Ti:sapphire amplifier output were focused into argon to produce phase-stable broadband MIR pulses in a well-focusable ring-shaped beam. The beam profile and spectrum of the MIR field are accurately reproduced with a simple calculation based on a four-wave mixing process. The ring-shaped pattern of the MIR beam originates from a dramatic confocal-parameter mismatch between the MIR field and the laser beams.

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References and links
Filamentation of powerful ultrashort laser pulses in gases [1–3] is one of the most interesting phenomena in nonlinear optics. The balance between self-focusing and plasma self-defocusing makes the pulse propagate much longer than the Rayleigh range with a very high intensity. It results in a dramatic enhancement of nonlinear processes occurring in the filamentation zone. This phenomenon enables high intensity pulse compression and efficient nonlinear wavelength conversion with gas media [2, 3].

Enhanced nonlinear-optical processes in laser-induced filaments suggest a new strategy for the generation of ultrashort pulses of long-wavelength radiation [4–11]. In particular, ultra-broadband MIR pulse generation is one of the most attractive applications of the filamentation effect. Such MIR pulses with more than one octave at full width at half maximum are very attractive to be applied for molecular spectroscopy, e.g. two-dimensional infrared spectroscopy. The MIR pulse generation through filamentation was firstly demonstrated in 2007 [7], and the technique was followed and slightly modified by several groups [8–11]. However, the precise characterization of the beam profile and pulse shape of the generated MIR pulses has not been reported so far.

In this paper, we report the detailed characterization of sub-cycle MIR pulse generation through a four-wave mixing (FWM) process in a filament. A full characterization of the beam profile and pulse shape of the conically emitted MIR field shows that this field was linearly polarized, well focusable, and its pulse duration was measured as 7.4 fs, which is much shorter than the single-cycle period of the center wavelength (3.9 μm) of the pulse. The spatial and temporal coherence of the generated MIR pulse is so high that the light source may result in a dramatic improvement in general MIR spectroscopy. Our simple numerical simulation accurately reproduced the beam profile and the spectrum of the MIR pulse.

The experimental setup is shown in Fig. 1. The light source was based on a Ti:sapphire multi-pass amplifier system (800 nm, 25 fs, 0.9 mJ at 1 kHz, Femtopower compactPro, FEMTOLASERS). The second harmonic (SH, ω2, 25 μJ) and the fundamental (ω1, 675 μJ) were
Fig. 1. Schematic of the system. Shaded region was purged with argon at atmospheric pressure. BS: beam splitter (5% reflection), BBO: $\beta$-BaB$_2$O$_4$ crystal (Type 1, $\theta = 29^\circ$, $t = 0.1$ mm), D: dichroic mirror, P: periscope, CM1: $r = 1$ m concave mirror, CM2: $r = 0.5$ m concave mirror, MH: aluminium-coated mirror with a hole ($\phi = 7$ mm), OAP: aluminium-coated off-axis parabolic mirror, BF: bandpass filter for 335-610 nm (FGB37, Thorlabs), OMA: spectrometer for ultraviolet region.

Fig. 2. (a) A typical spectrum of the mid-infrared pulse generated through filamentation in argon (filled curve). Sharp dips at 2400 cm$^{-1}$ and at around 1600 cm$^{-1}$ are due to absorption of residual carbon dioxide and water vapor, respectively. The four-wave mixing spectra calculated with the measured fundamental and the SH spectra before (dotted curve) and after (dashed curve) the filament. (b) Fundamental spectrum measured before and after the filament (dotted and filled curves, respectively). The spectrum did not change when the delay time between the fundamental and the SH pulses was adjusted. (c) SH spectra when the delay time between the fundamental and the SH pulses was not adjusted (solid curve) and was adjusted (filled curve).

spatially and temporally overlapped and focused into argon by a concave mirror ($r = -1$ m), generating a bright filament with a length of $\sim 3$ cm around the beam focus. This filament generated a MIR pulse ($\omega_0$) through a FWM process ($\omega_1 + \omega_1 - \omega_2 \rightarrow \omega_0$). The energy of this MIR pulse was measured as $\sim 250$ nJ by using a pyroelectric detector (J-10MB-LE, Coherent). With this energy level, it is possible to apply the pulses for the nonlinear spectroscopy of condensed matter. The pulse-to-pulse intensity fluctuation was about 2.5% rms.

The spectrum of the MIR pulse was measured with a home-built Fourier transform spectrometer. The measured spectrum is shown as a filled curve in Fig. 2(a). The broadband spectrum, which spread over the whole MIR region (500-5000 cm$^{-1}$), was due to the weak dispersion of the medium, with the phase-matching length ($0.23$ cm, evaluated including the effect of plasma...
Fig. 3. (a) Experimental and (b) simulated radial intensity distributions 250 mm after the generation point (on CM2 in Fig. 1) of the MIR pulses. (c) The intensity distribution at the focal point of the MIR pulse focused with a concave mirror (r=2 m).

and nonlinear refractive index [12]) exceeding the diffraction length of the IR beam (0.12 cm). Spectra of the fundamental and SH used for MIR generation are also shown in Figs. 2(b) and 2(c), respectively. The fundamental spectrum showed significant ionization-induced blue shift and the pulse was self-compressed down to 15 fs, whereas the SH spectrum was strongly broadened by cross-phase modulation. The MIR spectrum was compared with simple convolutions with the fundamental \(I_1(\omega)\) and SHG \(I_2(\omega)\) spectra \(\int d\omega' d\omega'' I_1(\omega') I_1(\omega'') I_2(\omega' + \omega'' - \omega)\) before (dotted line) and after (dashed line) the filament as is shown in Fig. 2(a). The MIR spectrum is well explained with the convolution of the spectra after the filament. This means that the blue shift and broadening of the input spectra in the filament are the key processes to generate shorter wavelength than terahertz. Terahertz wave generated due to the tunneling current [10], was much weaker than the signal in the MIR. According to our numerical simulations, the electron density achieved in our experiments was about \(3 \times 10^{17} \text{ cm}^{-3}\).

The beam profile of the MIR beam after ZnSe and Si filters measured with a pyroelectric camera (Pyrocam III, Spiricon) is shown in Fig. 3(a). The shape of the beam was ring and the angle of the cone was estimated to be about \(3^\circ\). Some asymmetric shape and distortion from ideal ring pattern comes from residual pulse front tilt and/or astigmatism of the light source. The generated MIR pulse has basically pure one-direction linear polarization (>40:1) in the entire cross-section of the beam as the input pulses, which fact was confirmed with a wire grid polarizer (NT62-774, Edmund Optics). We compare the experimental result with FWM-beam analysis after Ref. [13], based on a straightforward integration of the FWM response over the beam overlap region. As can be seen from Fig. 3(b), the simple approach provides an accurate agreement with the experimental result. It confirms that the ring-shaped beam profile originates from a dramatic confocal-parameter mismatch between the MIR field and the laser beams. Additionally, the \(~12\) mm diameter beam was focused down to 1.0 mm with a \(r=2\) m concave mirror, indicating a reasonable focusability for a ring shaped spatial mode. The beam profile at the focal point is shown in Fig. 3(c). Although the beam may contain some angular dispersion as was reported in Ref. [11], the dispersion is basically radially symmetric, and thus does not significantly deteriorate the good focusability of our MIR beam.

In order to quantitatively evaluate the temporal shape of the generated MIR pulse, we measured cross-correlation frequency resolved optical gating (XFROG) [14]. We used argon again as a nonlinear medium and used FWM process \(\omega_1 + \omega_1 - \omega \rightarrow \omega_2\) as a nonlinear interaction between the test pulse (MIR pulse, \(E_{\text{test}}(t)\)) and the reference pulse (\(E_{\text{ref}}(t)\)). The scheme is free from spectral filtering caused by phase matching condition in the nonlinear interaction. The system for the XFROG measurement is also shown in Fig. 1. Small portion (~1 \(\mu\)J) of the fundamental 25-fs pulse was used as a reference pulse. The reference pulse and the MIR
pulse (test pulse) were combined through a mirror with a hole and focused into argon with an aluminium-coated parabolic mirror ($f = 50$ mm). Generated blue spectra (centered around 440 nm) were measured with a spectrometer (USB2000+, OceanOptics) by scanning the delay time ($\tau$) between the reference pulse and the MIR test pulse. The reference pulse was independently characterized with SHG-FROG, and the result was used for retrieving the MIR pulse.

The measured and retrieved XFROG traces are shown in Fig. 4(a) and 4(b). The main feature of the trace indicates that the residual chirp of the test pulse is very small. The FROG error was 0.0009 with $256 \times 256$ grid. The retrieved time and frequency domain pictures are shown in Fig. 4(c) and (d), respectively. The pulse width is estimated to be 7.4 fs which is 0.57 cycles for 3.9 $\mu$m carrier wavelength. The retrieved spectrum was nearly identical to the spectrum measured with the Fourier-transform spectrometer, as is shown in Fig. 4(d). This indicates that the whole MIR spectral components were well focused and overlapped with the fundamental beam. Even the fine structure due to absorption line of the residual carbon dioxide was retrieved.

In theory, the carrier-envelope phase (CEP) of the generated MIR pulse is passively stabilized in the present scheme [7,15]. This feature makes the scheme highly attractive since CEP is very important physical property of sub-single cycle pulses. However, it is important to check the CEP stability experimentally since the fluctuation of the delay between the fundamental and SH pulses and some noise of the input pulse can affect the CEP stability [16–18].

The CEP stability measurement of the MIR pulses was carried out by measuring the interference between the SH of the reference pulse ($E_{\text{SHG}}(t)$) and the XFROG signal ($E_{\text{sig}}(t)$). The interference signal is explained as a cross-term of $|E_{\text{SHG}}(t) + E_{\text{sig}}(t)|^2$, namely, $E_{\text{SHG}}(t)E_{\text{sig}}(t) = E_{\text{SHG}}(t)E_{\text{ref}}^*(t)E_{\text{test}}(t)$. The phase of the interference signal is written as $-\phi_{\text{SHG}} + \phi_{\text{sig}} = -2\phi_{\text{ref}} + 2\phi_{\text{ref}} - \phi_{\text{test}} = -\phi_{\text{test}}$, where $\phi$ denotes CEP of $E_{\text{ref}}$ for each subscript. From the equation, it is clear that the phase drift of the interference signal reflects that of the CEP of the MIR test pulse. This scheme is essentially the same as that described in Ref. [19].

In the experiment, a barium borate crystal (BBO, $\theta = 29^\circ$, $t = 50$ $\mu$m) on a 2-mm thick fused
silica substrate was inserted behind the focus where the XFROG signal was generated. The crystal was placed in a way that the pulses enter from the back side, i.e., from the substrate side. This way, the XFROG signal is delayed by $\sim 300$ fs relative to the reference pulse due to the group delay difference in the substrate before hitting the BBO crystal. A calcite polarizer was inserted behind the crystal to optimize the intensity ratio between the SH and XFROG signals. In Fig. 5(a) the interference fringe is shown. The fringe spacing was about 3 THz which corresponds to the expected delay between the reference pulse and the XFROG signal. The phase of the fringe was reasonably stable for hours without any feedback loop. The instability of the phase was measured as 257 mrad rms with 100 shots (shown in Fig. 5(b)).

Due to the long wavelength cut off of the SH spectrum, the fringes were clear up to $\sim 415$ nm, which corresponded to $\sim 900$ cm$^{-1}$ of the MIR pulse. Although it is rather tiny component of the whole spectrum, it is safe to say that the fringe corresponds to the CEP of the pulse since the spectral phase retrieved from the XFROG results were well connected and has no phase jump in the region from 500 cm$^{-1}$ to 5000 cm$^{-1}$.

To demonstrate control of the CEP, the distance between fundamental and SH ($X$ in Fig. 1) was iteratively scanned with a 5-nm resolution feedback loop translation stage and the fringe was monitored (Fig. 5(c)). The fringe changes with a period of $\sim 400$ nm, which is similar situation as Refs. [19] and [20]. It was easy to control the CEP from 0 to $2\pi$ by changing the delay by 4 $\mu$m.

In conclusion, ultrabroadband coherent MIR spectrum which covers the entire MIR region was generated through conical emission from two-color filamentation. In our experiments, MIR pulses as short as 7.4 fs were generated, which corresponds to nearly half cycles of 3.9 $\mu$m center wavelength. We have revealed that the ring-shaped pattern of the MIR beam originates from a dramatic confocal-parameter mismatch between the MIR field and the laser beams.

The light source has a potential to change the situation of traditional MIR spectroscopy dramatically. For example, the coherent broadband MIR light source enables us to obtain absorption spectra through entire MIR region by single-shot with chirped pulse up-conversion technique [21, 22]. The reasonable quality of the spatial mode can be useful for efficient MIR microscope imaging combined with the up-conversion technique. Multi-dimensional spectroscopy for entire MIR region to monitor vibrational coupling among very different vibrational modes can be realized with the light source.

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