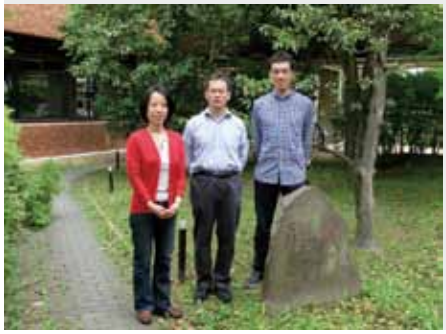
Ultrafast Laser Science

Laser Research Center for Molecular Science
Division of Advanced Laser Development



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Speed of ultrafast energy transfer from light to molecules (*i.e.* primary processes of photosynthesis, photoisomerization in visual pigments, *etc.*) is on the order of femtosecond (10^{-15} s). In our laboratory, we develop cutting edge lasers for such ultrafast molecular science, namely, femtosecond or attosecond (10^{-18} s) ultrashort pulse lasers.

For example, arbitrary waveform synthesis can be perf-ormed with simultaneous generation of femtosecond light pulses in various wavelength regions and superimposition of them with precisely controlled phases.

We would like to develop such advanced light control technology, which can push forward the research on ultrafast photochemical reactions.

1. Generation of Phase-Stable Half-Cycle Mid-Infrared Pulses through Filamentation in Gases[1)]

Filamentation of powerful ultrashort laser pulses in gases 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 fila-mentation zone. This phenomenon enables high intensity pulse compression and efficient nonlinear wavelength conversion with gas media.

Enhanced nonlinear-optical processes in laser-induced filaments suggest a new strategy for the generation of ultra-short pulses of long-wavelength radiation. Ultrabroadband mid-infrared (MIR, 3–20 μm) pulse generation through fila-mentation in air was firstly demonstrated in 2007,[2)] and the technique was followed by several groups. 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.

Here we report the latest progress of the ultrabroadband MIR pulse generation through filamentation. By using argon gas as the nonlinear medium and purging the optical path for the MIR pulse with the argon gas, a long tail of the pulse due to free induction decay of atmospheric carbon dioxide and water vapor disappeared, and it was possible to achieve gen-eration of high contrast MIR pulses. Full characterization of the pulse shape of the MIR field indicates that its pulse duration was 7.4 fs, which is about half-cycle period of the center wavelength (3.9 μm) of the pulse.

The experimental setup is shown in Figure 1. The light source was based on a Ti:Sapphire multi-pass amplifier system (Femtolasers, 800 nm, 25 fs, 0.9 mJ at 1 kHz). The second harmonic (ω_2, 25 μJ) and fundamental (ω_1, 675 μJ) pulses were spatially and temporally overlapped and focused into argon by a concave mirror (r = –1000 mm), generating a bright filament with a length of ~3 cm around the beam focus. This filament generated an MIR pulse (ω_0) through an ioni-zation-assisted wave mixing process ($\omega_1 + \omega_1 - \omega_2 \rightarrow \omega_0$). The energy of this MIR pulse was measured as ~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.

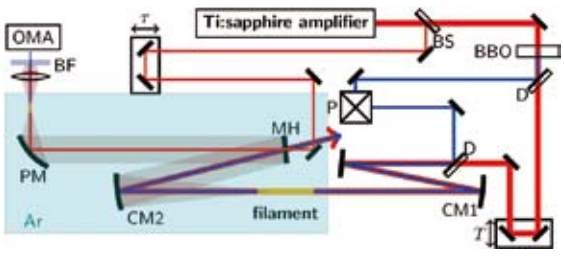

Figure 1. Schematic of the system. The shaded area was purged with argon. BS: beam splitter (5% reflection), BBO: β-BaB_2O_4 crystal (Type 1, θ = 29 deg, t = 0.1 mm), D: Dichroic mirror, P: Periscope, CM1: r = 1 m concave mirror, CM2: r = 0.5 m concave mirror, MH: Aluminium-coated mirror with a hole (ϕ = 7 mm), PM: Aluminium-coated parabolic mirror, BF: Bandpass filter for 335–610 nm (FGB37, Thorlabs), OMA: Spectrometer for ultraviolet region (USB2000+, OceanOptics).

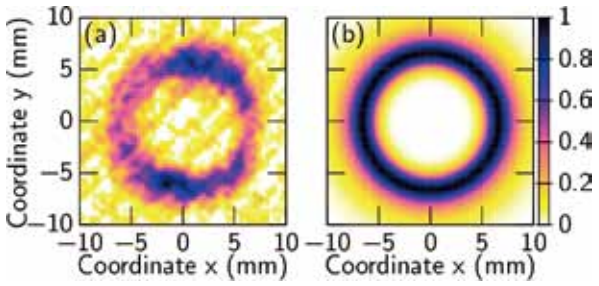

Figure 2. (a) Experimental and (b) simulated radial intensity distributions of the mid-infrared pulses.

The beam profile of the MIR beam after ZnSe and Si filters measured with a pyroelectric camera (Pyrocam III, Spiricon) is shown in Figure 2(a). The shape of the beam was ring and the angle of the cone was estimated to be about 3 deg. 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).

We compare the experimental result with FWM-beam analysis based on a straightforward integration of the FWM response over the beam overlap region. As can be seen from Figure 2(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. Although the beam may contain some angular dispersion, 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) We used argon again as a nonlinear medium and used four-wave mixing process ($\omega_1 + \omega_1 - \omega_0 \rightarrow \omega_2$) as a nonlinear interaction between the test pulse (MIR pulse) and the reference pulse. 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 Figure 1. Small portion (~2 μJ) 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 = 150 mm). Generated blue spectra (centered around 440 nm) were measured with a spectrometer (USB 2000+, OceanOptics) by scanning the delay time (τ in Figure 1) between the reference pulse and the MIR test pulse.

The measured and the retrieved XFROG traces are shown

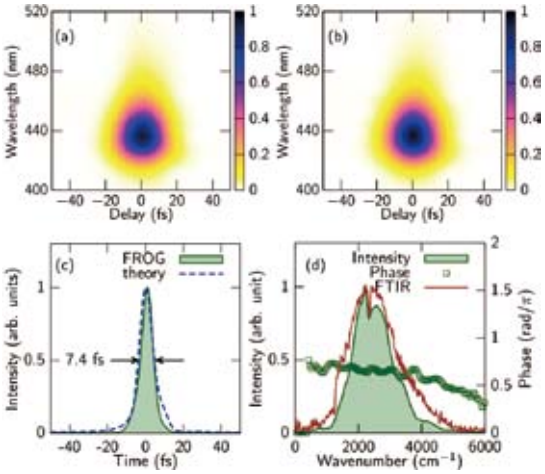

Figure 3. (a) Experimental and (b) retrieved XFROG traces. The retrieved pulse in (c) time and (d) frequency domain. The temporal profile from the 3D numerical simulation and the spectrum measured with Fourier transform spectrometer are also shown. Measured XFROG trace.

in Figure 2(a) and (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 × 256 grid. The retrieved time and frequency domain pictures are shown in Figure 2(c) and (d), respectively. The pulse width is estimated to be 7.4 fs, which is 0.57 cycles for 3.9 μm carrier wavelength. The retrieved spectrum is spread over whole MIR region (500–5000 cm^{-1}). The broadness of the spectrum was due to the weak dispersion of the medium, with the phase-matching length exceeding the length of the filament for all the MIR spectral components observed in the experiments.

In conclusion, ultrabroadband coherent MIR spectrum which covers the entire MIR region was generated through two-color filamentation. Due to the spatial and temporal quality, ultrashort MIR pulses generated in two-color filaments are ideal for numerous applications. In our experiments, MIR pulses as short as 7.4 fs were generated, which corresponds to about a half-cycle of 3.9 μm center wavelength.

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. 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.

References

1) T. Fuji and Y. Nomura, *21th International Laser Physics Workshop (LPHYS'12)*, 5.2.1 Calgary, July 23–27 (2012). (invited talk)

2) T. Fuji and T. Suzuki, *Opt. Lett.* **32**, 3330–3332 (2007).