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A Source for Ultrafast Continuum Infrared and Terahertz Radiation

Poul B. Petersen,1,2* and Andrei Tokmakoff2

1Department of Chemistry and Chemical Biology, Cornell University, 122 Baker Laboratory, Ithaca, NY 14853-1301, USA

2Department of Chemistry, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA 02139, USA

*Corresponding author: pbp33@cornell.edu

A compact and stable method for generating high intensity linearly polarized continuum mid-infrared and terahertz light using ultrafast femtosecond (fs) laser pulses is demonstrated. Continuous light generation from <400 cm⁻¹ (12 THz, 25 μm) to >3300 cm⁻¹ (100 THz, 3 μm) in a sub-100 femtosecond laser pulse is facilitated by nonlinear mixing of the fundamental, second harmonic and third harmonic of a ultrafast amplified laser source through filamentation in air. Including the third harmonic in the mixing scheme leads to a tenfold increase in the generated IR power. The compact optical configuration utilizing a delay plate in a collinear geometry serves to simplify alignment and increase stability making it a very practical source for transient IR spectroscopy.

OCIS codes: 190.4380, 260.3060, 320.6629, 320.7160
Continuum generation in the visible region has provided a powerful and readily available broadband source for time-and frequency resolved optical spectroscopy. Continuum is obtained by focusing an ultrafast laser pulse into a transparent nonlinear medium leading to self-phase modulation and other nonlinear processes. Analogous methods have been used to generate THz pulses in the single cycle regime.[1-5] Recently, broad-band single-cycle THz radiation has been generated through filamentation by simultaneously focusing the fundamental and second harmonic of an ultrafast laser pulse in air.[6-13] The mechanism of this latter process is described alternatively as four-wave optical rectification and tunnel ionization. For these processes the bandwidth of the generated continuum radiation is dictated by the bandwidth of the ultrafast laser source driving the filamentation. By using a 25 fs laser source at 800 nm and carefully controlling the amount of dispersion in the experimental setup, the frequency range of the continuum generation was recently extended to the mid-IR.[13]

Here we present experimental developments that further enhances the generation of ultrafast continuum mid-IR laser pulses to make it a practical source for ultrafast transient IR laser spectroscopy. The first experimental development is to include the third harmonic in the nonlinear optical mixing scheme leading to a much higher power of the generated continuum pulse. The power increase depends on the focal length of the mirror used to create the filamentation and is a factor of 10 for the present configuration using a 5 cm focal length curved mirror. The second experimental modification is the use of a delay plate to obtain temporal overlap in an all collinear geometry rather than using beam splitters and delay stages. This leads to a much more compact and simplified setup that is inherently very stable since all beams pass through the same optical components. Comparison of the $\omega/2\omega$ and the $\omega/2\omega/3\omega$ schemes are easily done by inserting or removing the tripling crystal and optimizing the waveplate angle with
minor adjustments of the delay plate. Using a 400 μJ, 35 fs laser source at 800 nm, the generated continuum pulse ranges from <400 to >3300 cm⁻¹ and is strong enough to saturate a liquid nitrogen cooled MCT array detector. In our measurements, the lower frequency limited is set by the detector response.

The experimental configuration for the ω/2ω/3ω scheme is illustrated in Figure 1. First 400 nm light was generated by type I frequency doubling the collimated 800 nm beam in a 200 μm thick BBO crystal cut at 29.3°. Third harmonic 267 nm light was generated under type I phase matching conditions in a 100 μm thick type I BBO crystal cut at 44.3°. A dual waveplate (14 λ at 400 nm and 6.5 λ at 800 nm) is used to tune the relative polarizations of the 800 and 400 nm pulses. Temporal walkoff in the doubling (39 fs), tripling (74 fs), and waveplate (92 fs) needs to be compensated for, but rather than using beam splitters and a delay stage to separate and recombine the 800 and 400 nm pulses, we adopted a much simpler design utilizing a delay plate.[1,4] The delay plate is in this case a 2 mm thick BBO cut at 66° that is inserted between the doubling crystal and the waveplate, where the 800 and 400 nm pulses propagate at orthogonal polarizations. When aligned with the slow (fast) axis along the 800 (400) nm polarization, the delay plate compensates for 195 fs temporal walkoff at normal incidence and is moderately angle tunable (8 fs/degree). After the tripling crystal, the fundamental, second and third harmonic are co-propagating and temporally overlapped. Filamentation in air and continuum generation is achieved by focusing the co-propagating beams by a 10 cm radius of curvature dielectric mirror (Layertec Art# 100600; this mirror is coated for 800 and 400 nm but also found to have a high reflectance at 267 nm) at a small angle to a spot size of ~20 μm. Maximum IR power was observed when the 800 nm polarization was at 45° with respect to the 400 nm leading to a 267 nm polarization perpendicular to the 400 nm light. Since all the beams
are co-propagating and pass through five common optics, this design is rugged, simple to set up, and the IR output has high stability (RMS<5%).

We have characterized the continuum mid-IR generation in terms of the spectral content, the power dependence, the polarization state, and the temporal profile. The spectrum of the mid-IR continuum pulse was characterized using a commercial FTIR spectrometer with a DTGS detector (Matson) and a liquid nitrogen cooled MCT array detector (IR Associates). The FTIR spectrometer is sensitive to the frequencies >400 cm\(^{-1}\), while the MCT array is optimized for < 8 \(\mu\)m (1250 cm\(^{-1}\)) wavelengths. The two spectra are compared to the radiation from the mid-IR lamp of the commercial FTIR in Figure 2a. Continuum generation is obtained from <400 (12 THz, 25 \(\mu\)m) to >3300 (100 THz, 3 \(\mu\)m) cm\(^{-1}\). Changing any of the input parameters, such as the polarization, time delay, and compression of the 800 nm pulse, strongly modulates the power of the continuum IR but the spectrum appears fairly invariant and must thus be dictated by the spectrum of the fundamental 800 nm beam. However, a slight broadening towards higher frequencies was observed in comparing the spectrum using the \(\omega/2\omega/3\omega\) scheme to the spectrum generated using the \(\omega/2\omega\) scheme.

Figure 2 further show the power dependence of the continuum mid-IR pulse as measured on the array detector. The pulse energy is estimated to be tens of nJ through comparison of the MCT response to a know laser source. Panel 2b display the integrated power of the continuum IR as a function of the total power of the 800 nm beam going into the setup. Above an initial threshold of 100 \(\mu\)J the continuum IR power increases according to a high order power law, and shows saturation behavior at higher pump fluence. The power-law coefficient is observed to depend on the wavelength: roughly \(E^5\) for frequencies in the center of the band (1900-2800 cm\(^{-1}\)) decreasing to roughly \(E^5\) for frequencies at the edge of the spectrum. Using a 2.5 cm focal length
curved mirror for filament generation leads to more efficient IR generation, however, the IR power saturates completely at 300 µJ incident 800 nm energy. Furthermore, the smaller focal length necessitates a larger angle between the incident and exciting beams and introduces a significant spatial chirp in the continuum IR. The spatial chirp was also observed using the 5 cm focal length, where using the 7.5 cm focal length mirror lead to lower power but almost negligible spatial chirp.

The polarization state of the continuum mid-IR was characterized by inserting a CaF₂ wire grid polarizer before the MCT array detector. At this point the IR continuum has been reflected off two 1” 90° uncoated gold off axis parabolas potentially leading to small changes in the polarization state. This change is estimated to be less than 3° and could explain that the observed polarization of a few degrees off from 45°. Figure 3 compare the IR intensity at selected wavelengths in the high frequency range (2200-3500 cm⁻¹) as a function of the analyzing polarizer angle. In the ω/2ω/3ω scheme, the polarization state of the continuum IR is linear at -45° within measurement error. A slight spectral variation is likely due to the fact that the dual waveplate is not perfect across the spectrum. In contrast, elliptically polarized light with a notable spectral dependence to the ellipticity was observed for the ω/2ω mixing scheme. This elliptical polarization could be due to a contribution to the continuum IR generated from the nonlinear crystals analogous to THz generation[7] as linear polarization was recently observed when care was taken to remove this contribution.[8,10]

These observed polarization characteristics follow the ZYYZ and ZYZY response of isotropic third-order nonlinear media for mixing processes such as ω_{3ω}=3ω−2ω−ω. Thus our findings for ω/2ω/3ω are consistent with the third-order optical rectification description put forward by Cook and Hochstrasser. However, the power dependence suggests either nonlinear
dependence of the plasma density or that even higher-order processes are contributing to the continuum generation in a perturbative treatment. Finally the temporal profile of the continuum mid-IR was characterized through both cross-correlation with a 267 nm pulse in CaF$_2$ and interferometric autocorrelation in the commercial FTIR. Multiphoton absorption of 267 nm light in CaF$_2$ generates free carriers which are probed in a frequency resolved manner by the continuum IR. The mid-infrared pulse length and group delay for frequencies between 1800-3550 cm$^{-1}$ is characterized by fitting the response to an Error function being the convolution of a step response function with two Gaussian pulse-profiles. The 267 nm light for the cross-correlation was generated independently with a 200 µm thick doubling crystal and a 300 µm thick tripling crystal. The 267 nm pulse duration is then limited by the walkoff in the tripling crystal, estimated to be 221 fs, and is the limiting factor in determining the pulse duration of the continuum IR. Figure 4a shows the fitted Gaussian pulse duration (FWHM of the intensity) and group delay obtained from fitting the cross-correlation as a function of the frequency of the continuum IR. The obtained pulse duration is in the range 100-150 fs and is thus limited by the relatively long 267 nm pulse. However, the group delay reveals a very small chirp on the pulse (~50 fs over the 1800-3500 cm$^{-1}$ frequency window). This relatively small chirp results from the low initial dispersion on the continuum mid-IR pulse and partly canceling dispersions in the Si-filter and CaF$_2$ window. The small dispersion on the continuum pulse means that we can obtain a good estimate of the continuum mid-IR pulse duration through the autocorrelation using the interferometer of a commercial FTIR (Fig. 4b). The field autocorrelation reveals a sub-2 cycle pulse with a FWHM of ~25 fs.

In conclusion, we have demonstrated the generation of ultrafast continuum mid-IR pulses using 800 nm and the first two harmonics through filamentation in air. The new scheme leads to
a much higher IR powers (a factor of 10 for the present setup) than what is generated by mixing only 800 and 400 nm pulses, and to a clean linear polarization. Furthermore, we used a compact setup utilizing a delay plate in an all collinear geometry rather than splitting up the pulses and using delay stages to ensure temporal overlap. This makes for a very stable and rugged design.

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References:


Figure 1. Experimental setup for the continuum mid-IR generation.

Figure 2. (a) Spectrum of continuum IR measured on MCT array (green) and FTIR (red) compared to the FTIR lamp spectrum (black). The blue and red bars at the top indicate the absorption bands of atmospheric H$_2$O and CO$_2$, respectively. (b) Power dependence of the integrated continuum IR generation as a function of 800 nm input energy.

Figure 3. Characterization of the polarization state of the continuum IR. Transmission of IR through an analyzing polarizer is measured as a function of polarizer rotation angle for the $\omega+2\omega+3\omega$ (a) and $\omega+2\omega$ (b) schemes.

Figure 4. Temporal characterization. (a) Gaussian intensity FWHM pulse duration (black) and group delay (red) cross-correlation of mid-IR with 267 nm light in CaF$_2$. (b) Interferometric autocorrelation measured in the commercial FTIR. Beats for delays >50 fs are primarily the free induction decay of atmospheric CO$_2$. 