Infrared extension of the supercontinuum generated by femtosecond terawatt laser pulses propagating in the atmosphere

J. Kasparian and R. Sauerbrey

Institut für Optik und Quantenelektronik, Max-Wien-Platz 1, 07743 Jena, Germany

D. Mondelain, S. Niedermeier, J. Yu, and J.-P. Wolf

Laboratoire de Spectrometrie Ionique et Moléculaire, Université Claude Bernard Lyon 1, Unité Mixte de Recherche 5579, Centre National de la Recherche Scientifique, F-69622 Villeurbanne Cedex, Lyon, France

Y.-B. André, M. Franco, B. Prade, S. Tzortzakis, and A. Mysyrowicz

Laboratoire d'Optique Appliquée, Unité Mixte de Recherche 7639, Centre National de la Recherche Scientifique Ecole Nationale Supérieure des Techniques Avancées—Ecole Polytechnique, Centre de l'Yvette, F-91761 Palaiseau Cedex, France

M. Rodriguez, H. Wille, and L. Wöste

Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany

Received July 5, 2000

We investigated the spectral behavior of a white-light continuum generated in air by 2-TW femtosecond laser pulses at 800 nm. The spectrum extends at least from 300 nm to 4.5 μm. From 1 to 1.6 μm the continuum's intensity increases strongly with the laser energy and depends on the initial chirp.

OCIS codes: 190.1900, 190.7110, 280.3640, 300.6340.

The propagation of high-peak-power laser pulses in transparent matter gives rise to strong nonlinear effects such as four-wave mixing,[1] stimulated Raman processes,[2] self-focusing,[3] and self-phase modulation (SPM),[4–6] which lead to strong modifications of the pulse characteristics. Self-focusing occurs because of the radial intensity variation in the laser beam, whereas SPM is due to the temporal variation of the laser intensity. Although both effects may be observed in conventional laser pulse propagation experiments, they dominate the behavior of ultrashort terawatt laser pulses. One of the most spectacular features observed is the formation of white-light filaments in air, so-called self-guided channels.[7,8] Self-focusing produces a large increase in rise of intensity in the filaments and permits multiphoton ionization of air and thus the formation of a low-density plasma, as demonstrated by electrical conductivity measurements.[9,10] Therefore self-focusing is balanced by diffraction as well as by refraction from the plasma, and thus the focused laser intensity is limited. The propagation is dynamically guided over distances much longer than the Rayleigh length, as much as several tens of meters,[8,11] with a diameter reported to be $\sim$100 μm.[7]

The spectral content of the white-light supercontinuum generated by high-power lasers has been a subject of interest since 1970.[1,12,13] However, because of limited peak power, experiments then were restricted to condensed media. Recently improvements in ultrashort lasers have permitted the results to be extended to gases. The spectral content of the supercontinuum generated by a 2-TW laser propagating in atmospheric-pressure rare gases has been measured in the visible and the UV, from 150 to 900 nm.[14] Until now, however, to our knowledge no experiment in the IR part of the supercontinuum generated in atmospheric-pressure gases has been performed.

In this Letter we report the measurement of the spectrum of the white-light continuum, particularly in the IR, up to 4.5 μm. The influence of the laser's initial power and chirp is also investigated. Besides the fundamental interest, that in measuring the IR part of the supercontinuum is stimulated by the potential application of the laser-induced continuum to lidar remote-sensing measurements.[15,16] The laser-induced continuum, as opposed to the traditional lidar technique,[17] allows simultaneous multispectral measurements to be made. This is especially interesting in the 3–3.5-μm IR band, where high-energy tunable laser pulses are difficult to produce and where a number of pollutant gases, in particular, volatile organic compounds have strong overlapping absorption bands.

In our experiments we used two state-of-the-art Ti:sapphire chirped-pulse amplification laser systems that had the following parameters. For system A (located at the École National Supérieure de Techniques Avancées): 60-mJ energy; 35-fs minimal pulse duration after the compressor (corresponding to 86-fs minimal pulse duration after the focusing lens); and beam diameter, 25 mm FWHM. The duration of the

0146-9592/00/181397-03$15.00/0

laser pulse was elongated by use of the grating compressor, thus producing chirped pulses. For system B (located at the Institut für Optkik und Quantenelektronic): 200-mJ energy; 100-fs minimal pulse duration after the compressor; and beam diameter, 35 mm FWHM. The energy was varied continuously. Both systems provided a peak power of ~2 TW at 800 nm (i.e., $\sim 10^{16}$ W/cm^2 if it was focused into a single filament), but, because the pulse durations were different, the intensity rise and fall times of the two systems were different.

We used several detection systems to cover the wavelength domain under investigation. For the visible spectral region an optical multichannel analyzer (Chromex 500 IS; $f/8$; spectral range, 400–1000 nm; grating, 150 lines/mm; resolution, 1.28 cm^{-1}; cooled; Si-ICCD-576, Princeton Instruments; 576 × 394 pixels) was used. In the IR, a prism spectrometer (Zeiss; 400 nm–2.7 μm; resolution, <5 nm) and a Perkin-Elmer double-pass prism spectrometer (focal length, 27 cm; LiF prism; transmission, up to 10 μm) were equipped with a germanium detector and a liquid-nitrogen-cooled InSb detector (1.5–5.6 μm, Hamamatsu). Additionally, IR interference filters (Corion) suppressed the fundamental wavelength of the laser beam.

In the experiment, the output of the laser was slightly focused with thin fused-silica lenses with 8- and 10-m focal lengths. We verified experimentally that, because of the lenses' large diameter (25 mm FWHM) and small thickness (3 mm), no continuum was generated in the lenses. Generation of the continuum occurred in the focal region, with the broadband continuum propagating farther with nearly the same divergence as the laser beam itself (see below). Power measurements before and after the focus showed that the continuum generation process caused no significant energy loss in the focus.

The spectral measurements took place at a total distance of ~30 m from the lens and hence after ~20 m of filament propagation. At this position the laser beam had a diameter of 20–25 cm. The angular pattern of white-light generation is beyond the scope of this Letter and is currently under investigation.[18] However, the white light is emitted mainly in the forward direction. Therefore all the spectral measurements were made in the forward direction, with an aluminum-coated mirror reflecting a small portion of the light to the entrance slit of the spectrometer used. The detection setup and the detectors were identical for both laser systems.

Figure 1 shows the spectral distribution of the white-light continuum generated in air by laser system A for a pulse duration of 115 fs after the focusing lens without chirp at the end of the compressor and for an 86-fs pulse duration. The spectrum was assembled from four single spectra (300–900 and 700–1800 nm and 1.5–2.7 and 1.5–4.5 μm) taken with four distinct detection systems with overlapping sensibility domains. The continuum band is very broad, extending at least to 4.5 μm.

In the IR, a region in which we know of no previous experiments in air, an almost exponential decay over 4 orders of magnitude up to 2.5 μm is observed. From 2.5 to 4.5 μm a slower decay is recorded, of 1 order of magnitude only. Above 4.5 μm, the spectral intensity was too low permit us to discriminate detector noise. Water absorption bands superimposed over a flat continuum spectrum are observed at 1.8 and 2.5 μm, suggesting that the use of a white-light continuum as a light source for spectroscopic remote sensing of the atmosphere[16] can be extended to the IR.

The dependence of the signal on the incident laser pulse energy was investigated with laser system B. As shown in the inset of Fig. 1, an initial factor-of-2 variation in intensity leads not to a variation in spectral shape in the 1–1.6-μm region to only to an overall decrease in efficiency by almost a factor of 1. Note that the conversion efficiency with laser system B, which provides shorter pulses, is 1 order of magnitude lower than with system A (Fig. 1), although both systems provide the same peak power of 2 TW.

The signal's dependence on the chirp (and thus the pulse duration) was investigated with laser system A. Figure 2 shows the continuum conversion efficiency at two wavelengths (1.7 and 3 μm, with 5-nm resolution) as a function of laser chirp. The optimal chirp setting depends on the emission wavelength to be optimized. Moreover, as shown in Fig. 1, the overall conversion efficiency in the near IR changes significantly when the chirp setting is changed.

SPM is generally believed to be the dominant process involved in continuum emission.[4–6,14,19] A simple calculation, with parameters that are typical of our experiments, and with a constant Gaussian beam, showed that SPM alone is in semiquantitative agreement with our experiments: (i) the calculated

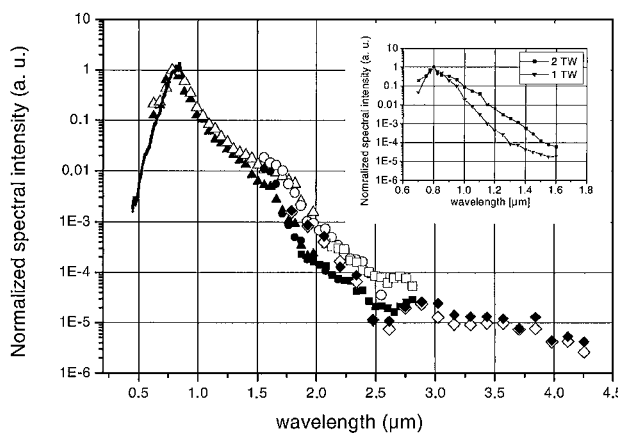

Fig. 1. Measured spectrum of the white-light continuum generated in the center of the beam by 2-TW laser pulses (laser system A). The results are shown for two different chirp settings that correspond to (i) an initial pulse duration of 35 fs without chirp after the compressor (filled symbols) and (ii) a 55-fs initial pulse duration with negative chirp after the compressor (open symbols). These curves are composed of 5 (4) distinct spectra from 5 (4) detection units, respectively, that correspond to the different symbol shapes for each curve. Inset, spectrum of the white-light continuum generated in the center of the beam by 100 fs pulses (laser system B) as a function of pulse power value (200 and 100 mJ for 2 and 1 TW, respectively). The two curves have the same normalization factor.

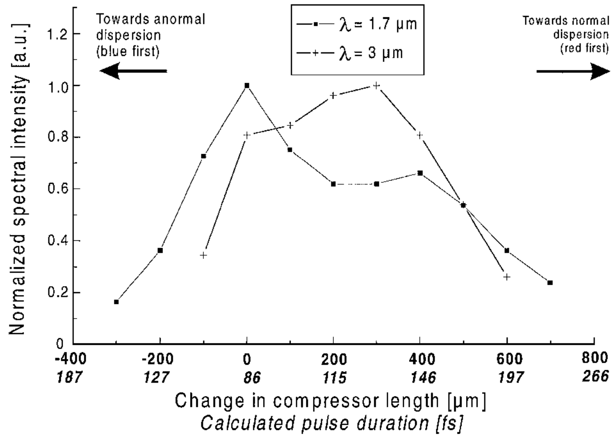

Fig. 2. Initial chirp dependence of the white-light continuum generated by laser system A at two distinct wavelengths (1.7 and 3 μm, with 5 nm resolution). The two curves have the same normalization. The chirp scale is shown as the change in the compressor length, with 0 set arbitrarily, to the chirp value of the curve with open symbols in Fig. 1. Numbers in italics are the corresponding calculated pulse lengths after the exit window and the lens.

spectral content of white light after 20-m propagation in a filament extends from a few hundred nanometers to at least 5 μm; (ii) laser system A, with shorter pulses, has a larger white-light generation efficiency than system B. This result supports the assumption that SPM is the main process in white-light continuum generation in air. However, a quantitative understanding of the continuum is still a subject for research and needs much more-complex discussion,[19,20] which is beyond the scope of this Letter. Understanding the influence of chirp will require more-detailed calculations.

For atmospheric applications the initial pulse chirp is also critical for the behavior of laser propagation. In particular, because of group-velocity dispersion in air, a negatively chirped pulse will recombine as an ultrashort pulse at a given distance away from the laser,[16,21] giving rise to localized white-light generation. The white-light pulse will also undergo group-velocity dispersion. However, because of tiny changes in the refractive index of air, the pulse broadening will be only 12 ps/km, as calculated with the Rank formula for a pulse ranging from 300 nm to 4.5 μm.

In conclusion, we have measured the wavelength dependence of the white-light continuum emitted from filaments generated in air by high-power femtosecond laser pulses up to 4.5 μm and down to 300 nm. The results show that the shape and the rate of rise in power of the excitation laser pulses have critical effects on conversion efficiency. As far as applications are concerned, a white-light continuum provides broadband pulses with a smooth spectrum up to at least 4.5 μm. It could therefore be used as an *in situ*–produced white lamp, permitting multispectral lidar measurements of atmospheric constituents to be expanded from the visible region[15,16] to the IR.

The authors acknowledge the help of the laser teams at the institutions at Palaiseau and Jena; of A. Reichmann, who provided the prism spectrometer; and of T. Töpfer, who provided valuable calibrations, tools, and some detectors. This study was done in the framework of the Teramobile project, a joint French–German project funded by the Centre National de la Recherche Scientifique and the Deutsche Forschungsgemeinschaft. R. Sauerbrey's e-mail address is sauerbrey@qe.physik.uni-jena.de.

References

1. R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. **24,** 584 (1970).
2. L. Smith, P. Liu, and M. Bloembergen, Phys. Rev. A **15,** 2396 (1977).
3. D. Strickland and P. B. Corkum, J. Opt. Soc. Am. B **11,** 492 (1994).
4. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. **24,** 592 (1970).
5. R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. **24,** 1217 (1970).
6. A. Brodeur and S. L. Chin, J. Opt. Soc. Am. B **16,** 637 (1999).
7. A. Braun, G. Korn, X. Liu, D. Du, J. Squier, and G. Mourou, Opt. Lett. **20,** 73 (1995).
8. E. T. J. Nibbering, P. F. Curley, G. Grillon, B. S. Prade, M. A. Franco, F. Salin, and A. Mysyrowicz, Opt. Lett. **21,** 62 (1996).
9. H. Schillinger and R. Sauerbrey, Appl. Phys. B **68,** 753 (1999).
10. S. Tzortzakis, M. Franco, Y.-B. Andre, A. Chiron, B. Lamouroux, B. Prade, and A. Mysyrowicz, Phys. Rev. E **60,** R3505 (1999).
11. M. Mlejnek, M. Kolesik, J. V. Moloney, and E. M. Wright, Opt. Lett. **23,** 2938 (1999).
12. W. Yu, R. R. Alfano, C. L. Sam, and R. J. Seymour, Opt. Commun. **14,** 344 (1975).
13. P. B. Corkum, P. P. Ho, R. R. Alfano, and J. T. Manassah, Opt. Lett. **10,** 624 (1985).
14. H. Nishioka, W. Odajima, K. Ueda, and H. Takuma, Opt. Lett. **20,** 2505 (1995).
15. L. Wöste, C. Wedekind, H. Wille, P. Rairoux, B. Stein, S. Nikolov, Chr. Werner, S. Niedermeier, H. Schillinger, and R. Sauerbrey, Laser Optoelektron. **29,** 51 (1997).
16. P. Rairoux, H. Schillinger, S. Niedermeier, M. Rodriguez, F. Ronneberger, R. Sauerbrey, B. Stein, D. Waite, C. Wedekind, H. Wille, and L. Wöste, "Remote sensing of the atmosphere using ultrashort laser pulses," Phys. B (to be published).
17. R. M. Measures, *Laser Remote Sensing—Fundamentals and Applications* (Wiley/Interscience, New York, 1984).
18. D. Mondelain, G. Ange, R. Volk, S. Niedermeier, J. Kasparian, J. Yu, R. Sauerbrey, and J. P. Wolf, Laboratoire Spectrométrie Ionique et Moléculaire, Université Claude Bernard Lyon 1, Unité Mixte de Recherche 5579, Centre National de la Recherche Scientifique, F-69622 Villeurbanne Cedex, Lyon, France, are preparing a manuscript to be called "Self-reflection-enhanced back scattering from white light filaments induced by high intensity femtosecond laser pulses propagating in air."
19. A. Gaeta, Phys. Rev. Lett. **84,** 3582 (2000).
20. I. Golub, Opt. Lett. **16,** 305 (1990).
21. J. Kasparian and J.-P. Wolf, Opt. Commun. **152,** 355 (1998).