Appl. Phys. B 77, 357–359 (2003)
DOI: 10.1007/s00340-003-1183-x

Applied Physics B
Lasers and Optics

Towards a supercontinuum-based infrared lidar

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Received: 31 January 2003/Revised version: 14 April 2003
Published online: 24 June 2003 •

ABSTRACT Lidar signals were obtained for the first time in the near-infrared using the supercontinuum generated by the terawatt femtosecond laser of the Teramobile system. A signal up to 4 km in altitude, in the band 1–1.7 μm, was collected using a 2 m astronomical telescope. We observed a 10-fold enhancement of the infrared signal backscattered from the atmosphere compared with that expected using a previously measured laboratory spectrum. This suggests a more efficient frequency conversion into the infrared (typically 7% into the 1–1.5 μm band) under long-distance propagation conditions.

PACS 42.65.Jx; 42.68.Wt; 42.65.Ky

1 Introduction

The Lidar (LIght Detection And Ranging) [1] technique has become a routinely employed tool for providing atmospheric pollution monitoring. The technique is, however, limited, especially for DIAL (DIfferential Absorption Lidar), to the measurement of one pollutant at a time, since the laser wavelength is tuned to an absorption line of the species being measured. Femtosecond lasers have recently raised hopes for the simultaneous detection of several pollutants [2, 3], based on white-light radiation through the physical process of supercontinuum generation (SCG).

SCG results from the non-linear propagation of femtosecond pulses with peak powers up to the terawatt level, even in a transparent and dilute medium such as air. The propagation of the pulse results in the creation of a self-induced guiding structure called a filament, interpreted as the manifestation of a dynamic equilibrium between Kerr lens focusing and ionization-induced defocusing (self-channeling [4]). Self-phase modulation (SPM) and possibly four-wave mixing occur, leading to spectral broadening around the frequency of the initial pulse. The supercontinuum spectral extent has been determined to reach from the UV to the near-infrared up to 4.5 μm [5].

Such bright thin filaments have been observed to develop over tens to hundreds of meters [6, 7], making them extremely promising for lidar applications in the visible spectral range, as shown by Wöste et al. [2, 3]. Extension of investigations to the infrared for possible atmospheric applications is desirable for the characterization of aerosol and gaseous species such as methane or volatile organic compounds (VOCs). However, lidar applications using the near-infrared side of the filament-generated supercontinuum remain a challenge, because of the rapid decrease of the infrared content of the supercontinuum and the λ^{-4} dependence of Rayleigh backscattering.

In this Letter, we report the detection of lidar signals in the near-infrared region up to 1.7 μm using the supercontinuum emitted from light filaments propagating in the atmosphere. This has been made possible by the combined operation of the container-embedded femtosecond terawatt laser of the Teramobile system [8] and an astronomic telescope. After suitable treatment, the detected backscattered band-integrated infrared radiation was compared with the spectrum measured in previous laboratory-scale experiments. Results show a strongly enhanced infrared spectrum from open-path propagation in the atmosphere, and therefore a more efficient frequency conversion than observed in laboratory-scale experiments.

2 Experimental set-up

The emitter of the lidar setup was the mobile femtosecond terawatt laser designed and built in the framework of the Teramobile project. This Ti:Sapphire-based laser chain delivered 290 mJ pulses, with a diameter of 9 cm, centered at 795 nm, at a repetition rate of 10 Hz. The initial pulse was slightly negatively chirped to compensate for the atmospheric group velocity dispersion, leading to an initial pulse dura-

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tionof 200 fs. The output 1.5 TW beam, sent vertically, was focused a few tens of meters high in the atmosphere through a telescope of adjustable focal length.

The receiver consisted of the 2 m primary mirror telescope [9] of the Thüringer Landessternwarte (TLS) observatory located 30 m from the laser. The Coudé configuration telescope had a field-of-view of 1.2 mrad (full aperture).

The collected light was imaged with an apparent aperture number of $f/92$ onto a liquid-nitrogen-cooled photomultiplier tube (PMT, Hamamatsu R 5509-72, sensibility range 300 nm–1.7 μm). To analyze the IR white-light lidar return, we recorded the signals in several spectral bands by using 4 filter sets: (i) no filter, (ii) 1–1.7 μm by use of a long-pass filter (Corion), (iii) 1.2–1.7 μm by use of a combination of two 1 mm thick Schott glass filters UG7 and VG12, and (iv) 1.5–1.7 μm by use of a long-pass filter (Corion). The signal was acquired and averaged over 16 shots on a digital oscilloscope, triggered by the laser pulses. The chirp in the laser compressor and the focus of the sending telescope were optimized to give the maximal detected signal at high altitude. The astronomical telescope was pointed towards the laser beam at about 4 km altitude, at the bottom of a cloud layer.

3 Results and discussion

Infrared backscattered signals could be detected with all filter sets used, even for the 1.5–1.7 μm range (Fig. 1). This is, to our knowledge, the first observation of an infrared lidar signal based on supercontinuum generation.

Because of the very limited spectral resolution of the measurement, we compared the actual lidar data with a simulation of the lidar signal that would have been observed if the initial IR spectrum was the same as measured in the laboratory [5]. The simulation also assumed that the previously measured supercontinuum spectrum was generated at low altitude and then propagated linearly over the 8 km forward and return path to the cloud. Since the sky was clear from the ground to the cloud, we neglected any interaction between the laser beam and low-altitude aerosols. We took into account

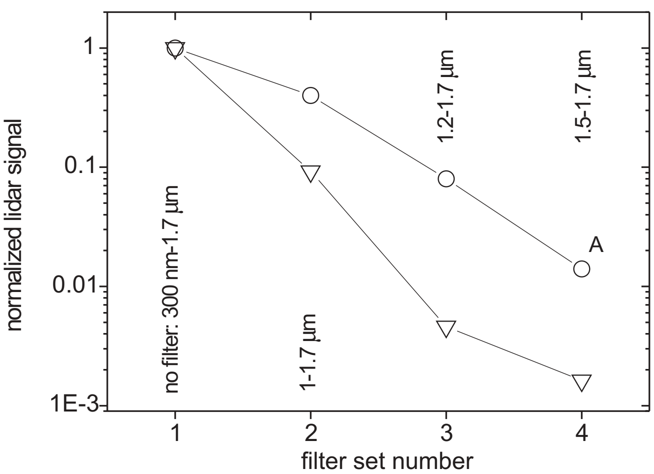

FIGURE 1 IR lidar signal for each filter set (*circles*) and expected values calculated from the laboratory spectrum [5], using a C.1 cloud size distribution centered at $r_0 = 4\,\mu m$ (∇). Each curve is normalized to 1 at the fundamental wavelength (filter set 1)

the transmission functions of the diverse filter combinations that we used, the PMT spectral response, and the transmission of the (fully gaseous) atmosphere. This atmospheric transmission was calculated using HITRAN, assuming a mean value for the relative humidity (80%) and the temperature (270 K), both inferred from radiosonde data. The interaction with the cloud was modeled using Mie scattering calculations, and the contribution of multiple scattering was evaluated following a method derived from Bissonnette's multiple-scattering lidar equation (MSLE) [10]. This model only considers the forward and backward lobes of the angular Mie scattering pattern and approximates them by Gaussian curves, thus allowing a quasi-analytical calculation. In our calculations, the backward peak was modeled by a polynomial instead, in order to take into account the ripple structures in the backward direction due to large particles. Besides the density of the cloud, the MSLE requires the particle size distribution as an input parameter. From the altitude and opacity of the cloud, we considered as a first approach an altocumulus cloud of spherical water droplets with radii that follow a C.1 Deirmendjian size distribution [1]:

$$dN/dr = CN_0/r_0(r/r_0)^6 \exp(-6r/r_0)\,, \tag{1}$$

where N is the concentration, r is the particle radius, $r_0 = 4\,\mu m$ is the mode of the size distribution, $C = 388.8$ is a normalizing factor, and N_0 is the overall aerosol density, which was determined using a self-consistent method applied to the elastic lidar signal. More precisely, the applied procedure was as follows: (1) a first estimate $N_0^{(1)}$ was obtained from the backscattered signal, neglecting multiple scattering; (2) $N_0^{(1)}$ was used to compute the multiple scattering contribution; (3) this multiple scattering contribution was used to retrieve a second estimate $N_0^{(2)}$ from the measured lidar signal; (4) the procedure was iterated until convergence was achieved. After 4 iterations, this algorithm converged to a particle density of $N_0 = 1.2\,cm^{-3}$, corresponding to an aerosol extinction of $16.3\,km^{-1}$ at 800 nm, and a relative contribution of multiple scattering evaluated to be 30% at the fundamental laser wavelength, and decreasing to 15% at $\lambda = 2\,\mu m$. The retrieved extinction value corresponds to a dense altocumulus cloud. For all of the calculations, only the real part of the refractive index was taken into account, which is not expected to significantly affect the results, since we measured at the bottom of the cloud and the backscattering coefficient is only weakly modified by the absorption.

The simulated IR lidar signals calculated from laboratory spectral measurements lie much below the measured results (respectively ∇ and O symbols on Fig. 1), with a ratio of about one order of magnitude above 1.5 μm. This indicates that the frequency conversion into the spectral region 1–1.7 μm was significantly more efficient in our large-scale experiment than in previous laboratory measurements. This is due to the much longer interaction path in the atmosphere. Notice that differences between long-range propagation and focused geometries have also been observed in the visible: a plateau has been observed in the visible side of the spectrum emitted by filaments generated by slightly focused beams (see e.g. [3] and [7]), while a sharp decrease has been observed in the case of stronger focusing (such as e.g. in [5]). Moreover, the at-

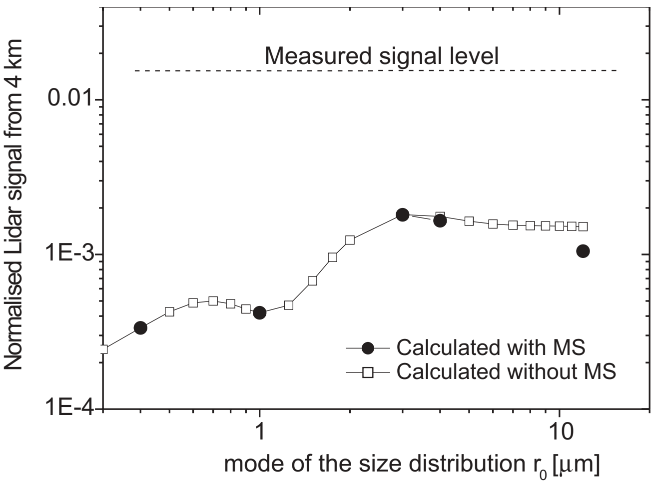

FIGURE 2 Influence of mode of the C.1 size distribution of the cloud on the calculated lidar intensity with filter set 4 (1.5–1.7 µm). Calculations both with and without multiple scattering are presented. The normalization is the same as in Fig. 1. The *horizontal dotted line* is the relative intensity of the measured lidar signal (point A of Fig. 1)

mosphere is clear below the cloud layer. Hence, no influence from low-altitude atmospheric aerosols on the SCG was expected in our experiment.

Since the cloud size distribution was set to a plausible value, we checked the influence of the mode r_0 of the particle size distribution on the retrieved intensity in a fixed wavelength range of filter set 4 (1.5–1.7 µm). For values of r_0 above 2 µm, the calculated intensity backscattered from the cloud exhibits a weak dependence and reaches a maximum for $r_0 = 3$ µm. Below 3 µm, it drops significantly as a consequence of the lower Mie scattering efficiency in the IR. Results of the calculations are presented in Fig. 2, both with and without taking multiple scattering into account. Even in the worst case situation ($r_0 = 3$ µm) the observed IR signal is 7 times larger than the signal expected from the laboratory spectrum [5]. Similar behavior was observed for the other filter sets. The same calculation was performed with and without taking multiple scattering into account, showing that the multiple scattering is negligible for size distributions centered below several microns, while for larger particles, its effect is an enhancement of the difference between the measured and calculated signals.

In order to get more insight into the spectral dependence of our lidar measurement, we assumed that the decrease of the supercontinuum spectral profile generated in the atmosphere follows an exponential decay on the IR side, as observed in the measurements under laboratory conditions [5]. By adjusting the corresponding exponential slope factor to the experimental data, we obtained a decrease of 1 decade per micron, instead of 3 decades as for the laboratory-scale spectrum. This corresponds to a 7% energy conversion efficiency into the 1–1.7 µm band, instead of 0.5% in the laboratory spectrum of [5]. This high efficiency conversion would make the IR part of the SCG a good candidate for many applications requiring ultrashort pulsed IR broadband sources, such as lidar or time-resolved spectroscopy.

4 Conclusion

Near-infrared lidar signals have been recorded from an altitude of up to 4 km, using the broadband white light generated from filaments propagating in the atmosphere. The signal measured for infrared wavelengths is about ten times stronger than expected from simulations from previous laboratory experiments [5]. This result could be attributed to the longer interaction path that results from a weaker focusing. Signal extension to the mid-infrared spectral range can be valuable for the detection and identification of atmospheric pollutants (particularly VOCs) that display characteristic absorption bands in this spectral region. The knowledge of the white-light spectral density is a key feature, as it constitutes the background spectrum of potential altitude-resolved absorption spectra for multi-pollutant detection. This work yields interesting insights for the application of supercontinuum-based LIDAR systems.

ACKNOWLEDGEMENTS We acknowledge the support of the Teramobile project, jointly funded by the Centre National de la Recherche Scientifique (France) and the Deutsche Forschungsgemeinschaft (Germany). J.-P. Wolf acknowledges financial support from the Institut Universitaire de France. The Teramobile members would like to thank the technical staff of the Thüringer Landessternwarte Tautenburg, Bernd Furhmann, Jürgen Haupt, Axel Kirchhof, Michael Pluto, Jörg Schiller, and Johannes Winkler, for their assistance in installing the laser at TLS and helping to solve technical problems. We also thank Eike Guenther, Sylvio Klose, and Helmut Meusinger for useful discussions on conducting the observations with the 2 m telescope. The Teramobile web site is www.teramobile.org.

REFERENCES

1 R.M. Measures: *Laser Remote Sensing – Fundamental and Applications* (Wiley Interscience, New York 1984)
2 L. Wöste, C. Wedekind, H. Wille, P. Rairoux, B. Stein, S. Nikolov, C. Werner, S. Niedermeier, H. Schillinger, R. Sauerbrey: Laser Optoelektronik **29**, 51 (1997)
3 P. Rairoux, H. Schillinger, S. Niedermeier, M. Rodriguez, F. Ronneberger, R. Sauerbrey, B. Stein, D. Waite, C. Wedekind, H. Wille, L. Wöste: Appl. Phys. B **71**, 573 (2000)
4 A.L. Gaeta: Phys. Rev. Lett. **84**, 3582 (2000)
5 J. Kasparian, R. Sauerbrey, D. Mondelain, S. Niedermeier, J. Yu, J.-P. Wolf, Y.-B. André, M. Franco, B. Prade, S. Tzortzakis, A. Mysyrowicz, M. Rodriguez, H. Wille, L. Wöste: Opt. Lett. **25**, 1397 (2000)
6 B. La Fontaine, F. Vidal, Z. Jiang, C.Y. Chien, D. Comtois, A. Desparois, T.W. Johnson, J.-C. Kieffer, H. Pépin: Phys. Plasmas **6**, 1615 (1999)
7 T.J. Nibbering, P.F. Curley, G. Grillon, B.S. Prade, M.A. Franco, F. Salin, A. Mysyrowicz: Opt. Lett. **21**, 62 (1996)
8 H. Wille, M. Rodriguez, J. Kasparian, D. Mondelain, J. Yu, A. Mysyrowicz, R. Sauerbrey, J.P. Wolf, L. Wöste: EPJ-AP **20**, 3 (2002)
9 H. Lehmann: http://www.tls-tautenburg.de/telesc.html (2002)
10 L.R. Bissonnette: Appl. Opt. **35**, 6449 (1996)