Ultrashort Laser applications in Lidar and atmospheric sciences

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ABSTRACT

The propagation of ultrashort, ultra-intense laser pulses gives rise to strongly nonlinear processes. In particular, filamentation is observed, yielding a ionized, conducting plasma channel where white-light supercontinuum due to self-phase modulation occurs. This supercontinuum, extending from the UV to the IR, is a suitable "white laser" source for atmospheric remote sensing, and especially Lidar (Light Detection and Ranging). Recent significant results in this regard are presented, as well as lightning control using ultrashort laser pulses. The application of ultrashort-pulse lidar to aerosol monitoring is also discussed.

Keywords:Lidar, remote sensing, ultrafast lasers, ultrafast nonlinear optics, white-light generation, atmospheric propagation, aerosol detection, lightning control.

1. INTRODUCTION

Since many years lasers have been used in atmospheric diagnostics, particularly in remote sensing. Lidar (Light Detection And Ranging)[1] has become a powerful technique to monitor atmospheric parameters and has helped to understand a variety of atmospheric phenomena. In the field of laser research, on the other hand, the generation of ultra-short laser pulses made the decisive step forward with the development of the chirped pulse amplification (CPA) technique in 1985.[2],[3] It provides femtosecond pulses with peak power values exceeding 10^{12} W and enormous short-time intensities. Both aspects together suggest to evaluate if the application of high-intensity lasers, in combination with the Lidar technique, can lead to novel methods in atmospheric diagnostic. This was first tried by Wöste *et al.*[4] Later, Rairoux *et al.*[5] demonstrated the suitability of the white-light beam generated by a TW-laser for remote atmospheric spectroscopy. The outstanding new aspect about the femtosecond Lidar is that the air, which is a matter composed of gases and particles, i.e. aerosols, plays a double role. It is not only the object of analysis, but becomes a key element of the metrology itself. Extremely non-linear light-matter interactions promise new possibilities for atmospheric science, but, to be able to interpret the measurements correctly, they also require intensive investigations. Beside the importance for the development of the femtosecond lidar, those investigations are part of the current basic research in quantum optics.

In the following, we briefly describe the relevant properties of nonlinear propagation of ultra-intense pulses in air and show some related experimental results. All laser systems used in this work are titane-sapphire CPA systems which emit fs-pulses at a central wavelength of approximately 800 nm and with a spectral width of typically around 20 nm. The main focus lies on the creation of plasma filaments, caused by the so called self-guiding or self-channelling effect,[6] and the spectral broadening of the laser pulse, which is called supercontinuum generation. Those results have mainly been obtained in laboratories, but when dealing with TW-pulses the propagation range in which the interaction processes take place exceeds, by far, usual laboratory dimensions. Therefore long-range propagation experiments (in the order of kilometers) are needed, also because there is still a lack of numerical simulation models that allow to calculate the propagation with the needed precision in a reasonable speed. To overcome this limitation and, of course, to obtain the possibility to perform femtosecond lidar measurements at nearly any desired spot, we developed and built the first mobile terawatt laser system, the Teramobile. This autonomously operating laboratory, based on a standard freight container, has been described in detail elsewhere.[7] It contains all elements of the femtosecond lidar system, as shown schematically in Figure 1. Recent results obtained with the Teramobile are presented in Sections 3, 5 and part of 2.1.

Nevertheless, laboratory experiments still play a key role in our femtosecond lidar research. In Section 4 we shortly present some measurements of the interaction between intense laser pulses and particles. Under laboratory conditions scattered and emitted light from single tailored microdroplets (liquid aerosol) can be detected. Those experiments provide fundamental knowledge that is needed to gain information about atmospheric aerosols out of femtosecond lidar measurements.

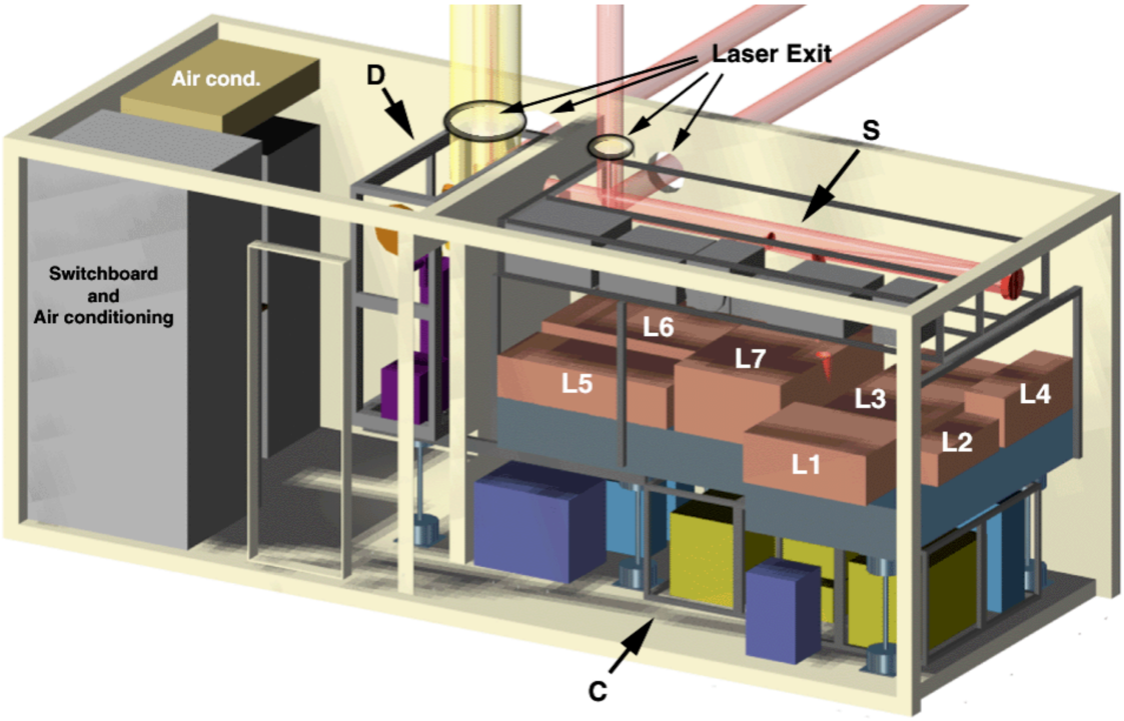


Figure 1. Schematic of the Teramobile mobile laser laboratory. L1-L7 : laser components, C: separately cooled box hosting the power supplies under the table, S: sending telescope with zooming capability, D: Lidar detection system.

2. NONLINEAR PROPAGATION OF HIGH POWER LASER PULSES IN AIR

At high laser intensities the optical Kerr effect, *i.e.* the intensity dependent variation of the refractive index, balances the beam diffraction, resulting in self-focusing of the beam. In air the critical power for self-focusing is approximately 2 GW. This theoretical value corresponds to a diffraction-limited gaussian beam. At this intensity, the self-focusing length starts to descend from infinity. If the pulse power exceeds this level, self-focusing leads to a further enhancement of the intensity and to a catastrophic collapse of the pulse at a non-linear focus. However, when the intensity approaches 10^{13} to 10^{14} W/cm^2 multiphoton ionization (MPI) starts. The appearance of an underdense plasma results in an intensity dependent reduction of the refractive index, defocusing the beam. This prevents the beam from collapsing into a plasma flash – as happens when the beam is focussed with a strong lens. Instead of that an equilibrium between Kerr focusing and plasma defocusing produces thin intense filamentsFigure 2. Propagation of such self-guiding filaments with a stable diameter has been observed over tens (up to hundreds) of meters.[8] The propagation of a TW pulse normally results in multi-filamentation. Inhomogeneities in the beam profile act as seeds for filaments. On the other hand, several experimental and theoretical works show quite constant values of the filament diameter (approximately 100 µm) and intensity (slightly above the MPI threshold).[6,9,10,11] Indeed it seems to be plausible that these are intrinsic properties of such filaments and that therefore the energy carried by one filament is limited. In the filaments, the high intensity yields an efficient self-phase modulation (SPM), which is the temporal counterpart of the Kerr effect: the time variation of the intensity results in a time variation of the refractive index, and hence a time dependent nonlinear phase, which results in the generation of new wavelengths, emitted as a conical emission. This effect, together with four wave mixing (FWM) and the group velocity dispersion (GVD), leads to strong modifications of the spectral characteristics and the temporal shape of the pulse.

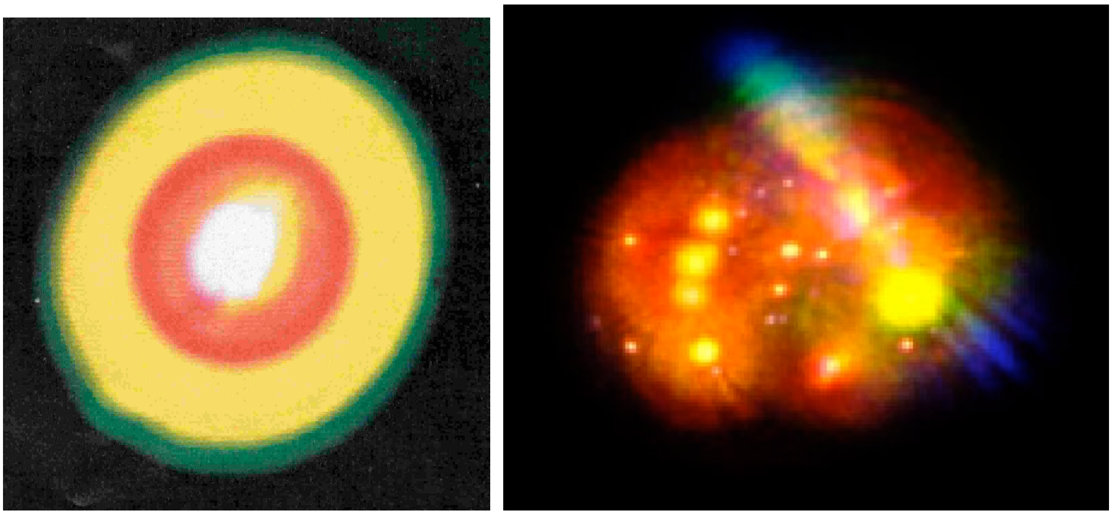


Figure 2. Beam image on screens. Left: conical emission from a single filament at a power slightly above the critical power; Right: multiple filamentation for a pulse energy much over the critical power

2.1 CONTROL OF THE FILAMENTATION

As can be observed in Figure 2, the multi-filamentation created by a TW laser is relatively stable from pulse to pulse, although the single filaments slightly fluctuate in space. The free propagation length after which the filaments start depends on the laser parameters, according to the mechanisms of self-focussing. The starting point of the zone where the light becomes intense enough to ionize the air can be chosen by use of focussing optics. With an initially collimated beam, one can observe a single filament or a narrow bundle of few parallel filaments, with a cone of the red rest light around it.

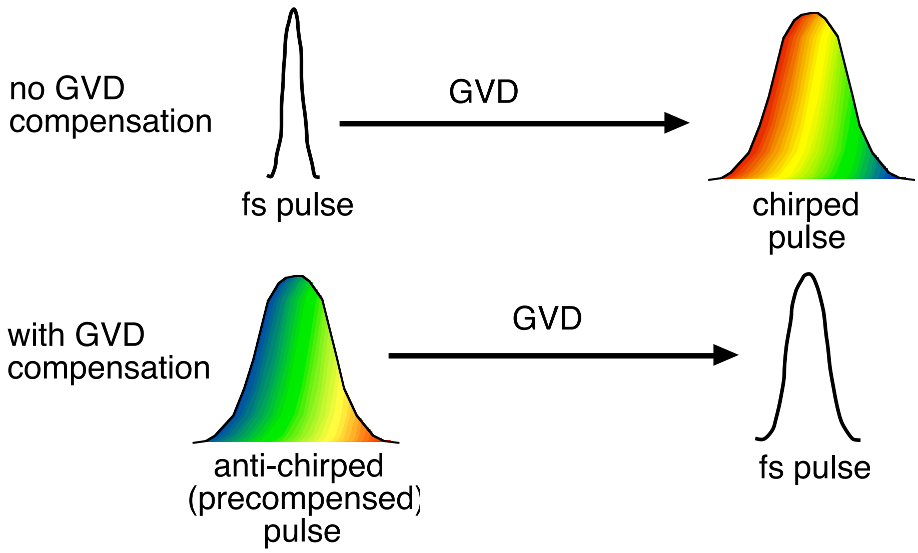


Figure 3. Principle of group velocity precompensation with an antichirp

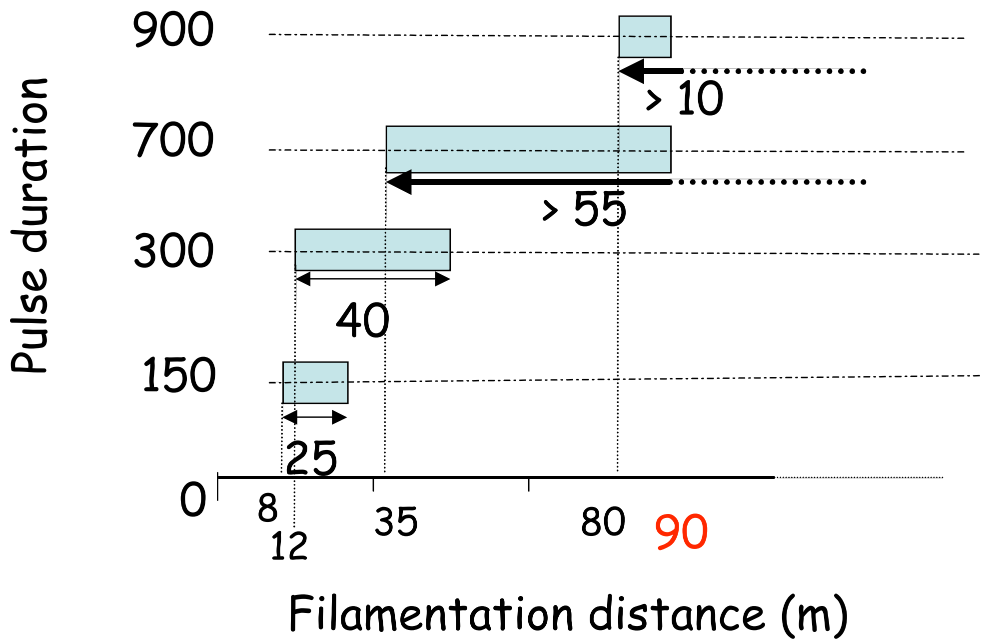


Figure 4. Chirp dependence of the filament position and length. The shortest pulse in the experiment was approx. 100 fs long. Here the value 150 fs stands for a slight positive chirp, the three upper durations represent the length of pulses with negative chirp.

One way to control the filamentation distance over long distances is to use the GVD in air. For that we tune the compressor setting of the CPA chain to give a linear chirp to the pulse, *i.e.* to disperse the spectral parts of the light over time. This makes the pulse longer, but with a so called negative chirp. The GVD recompresses this chirp during the propagation (Figure 3). Figure 4 shows results obtained with the Teramobile system on a range of 90 m. The measured effect is faster than expected from GVD, which is 1 fs/m for a 16 nm broad pulse. But, since the pulse power lies far above the self-focussing threshold for all pulse durations set in this experiment, the nonlinear optical effects have to be considered along the whole propagation. This is consistent with the fact that also a slightly positive chirped pulse undergoes self-channeling (pulse duration 150 fs in Figure 4). However, these results show that the chirp permits to control the filamentation distance.

A precise control of the filamentation is very important for Lidar as well as the other applications presented in the following sections. The determination of the range in which the filaments interact with molecules and particles is highly desired. Moreover this subject is closely linked to the investigation about the properties of the light that comes out of the filaments and their dependencies on the laser parameters.

2.2 PROPERTIES OF THE WHITE-LIGHT CONTINUUM

The creation of white light with lasers in condensed matter is well known since the early 1970s, but the supercontinuum generation (SCG) in gases[12] first became possible with the development of high-power lasers. In air the conditions for an efficient spectral broadening seem to be optimal inside filaments with the properties described in Section 2.1. The spectral content of the white light has been measured by Kasparian *et al.*[13] with a strong focusing, putting emphasis on the infrared part where many trace gases have their characteristic absorption lines. Light up to wavelength of over 4 µm could be detected (see Figure 5). This spectrum covers the absorption band of many pollutants of interest, opening the way to multipollutant Lidar measurements based on the white-light supercontinuum.

Spectra in the visible region have been obtained from Lidar experiments, *i.e.* measuring the backscattered light from the atmosphere in 1 km height.[5] In both laboratory and Lidar measurement, the shape of the spectrum depends on the pulse power and the chirp setting. Horizontal kilometer-range propagation experiments are planed to be performed with the Teramobile, in order to gain more information, *e.g.* how to obtain the maximal intensity in a given spectral range. Nevertheless, the already obtained data are important for the development of the white-light lidar, as described in Section 3. On the other hand, the Lidar experiments themselves contribute information about the properties of the filament emission.

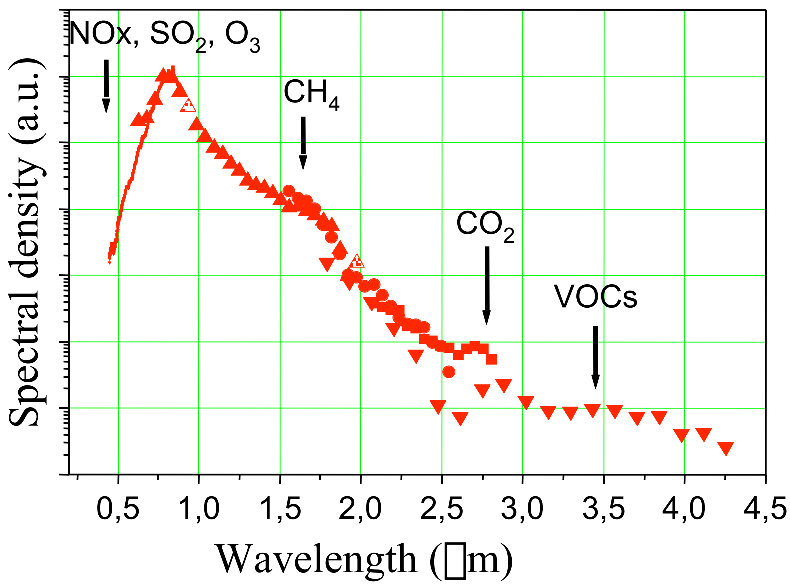


Figure 5. Measured spectra of 2-TW pulses after propagation of 30 m behind a 10-m lens.

3. FEMTOSECOND WHITE-LIGHT LIDAR

The main advantage of lidar over other remote sensing techniques, such as differential optical absorption spectroscopy (DOAS),[14] Fourier transform infrared spectroscopy (FTIR)[15] or satellite based spectroscopy,[16] is the high range resolution over long distances, which is achieved by the use of short-pulse lasers (typically a few nanoseconds in usual Lidar systems) and fast electronics to record the signal of the light backscattered by molecules and aerosols. However, standard Lidar systems are normally restricted to the detection of one trace gas at a time. Furthermore the number of the detectable species is limited by the availability of narrow-lined pulsed laser sources at suitable wavelengths, especially in the IR. DOAS and FTIR overcome this limitation by use of a wide spectral continuum of natural or artificial light sources, but they have their deficits in spatial resolution and choice.

A femtosecond white-light lidar can combine the advantages of both kinds of remote sensing techniques, adding a broadband spectral resolution to the 3D mapping capability of Lidar. This permits the simultaneous measurement of several compounds, even with overlaping spectra, as is the case for the volatile organic compounds. Moreover, since the whole spectrum can be acquired at once, the shot-to-shot fluctuations of the laser beam do not yield a systematic error as is the case for DIAL Lidar. Moreover, experiments have shown that the white light emitted from self-guided filaments is forward directed and propagates as a collimated beam over large distances.[4,17]. Moreover, Yu *et al.*[18] showed that, due to local laser-induced refractive index gradients, the backward supercontinuum emission is significantly enhanced, *i.e.* more white light is emitted towards the lidar detection system from the filaments than by elastic backscattering.

The results showed here were obtained with the Teramobile system. During the campaign, it was stationed 30 m away from the astronomical 2-m telescope of the Thüringer Landesternwarte in Tautenburg, Germany. The observatory was used as the lidar detection unit in two ways. With the telescope set to the Schmidt configuration images of the laser beam were taken with a field of view of 0.6°. In the Coudé configuration light backscattered from different heights was detected under a field of view of 1.2" and analyzed by a high resolution Échelle spectrograph.

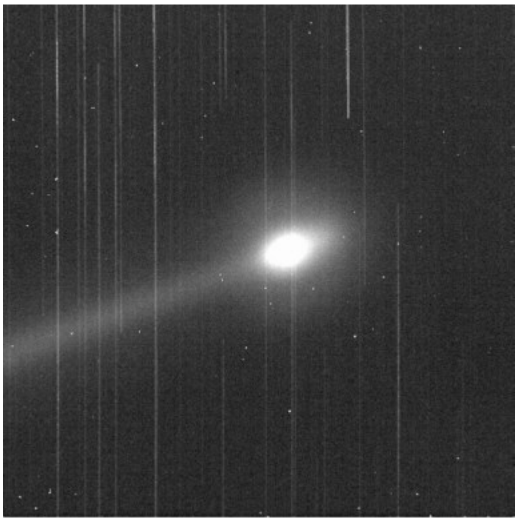
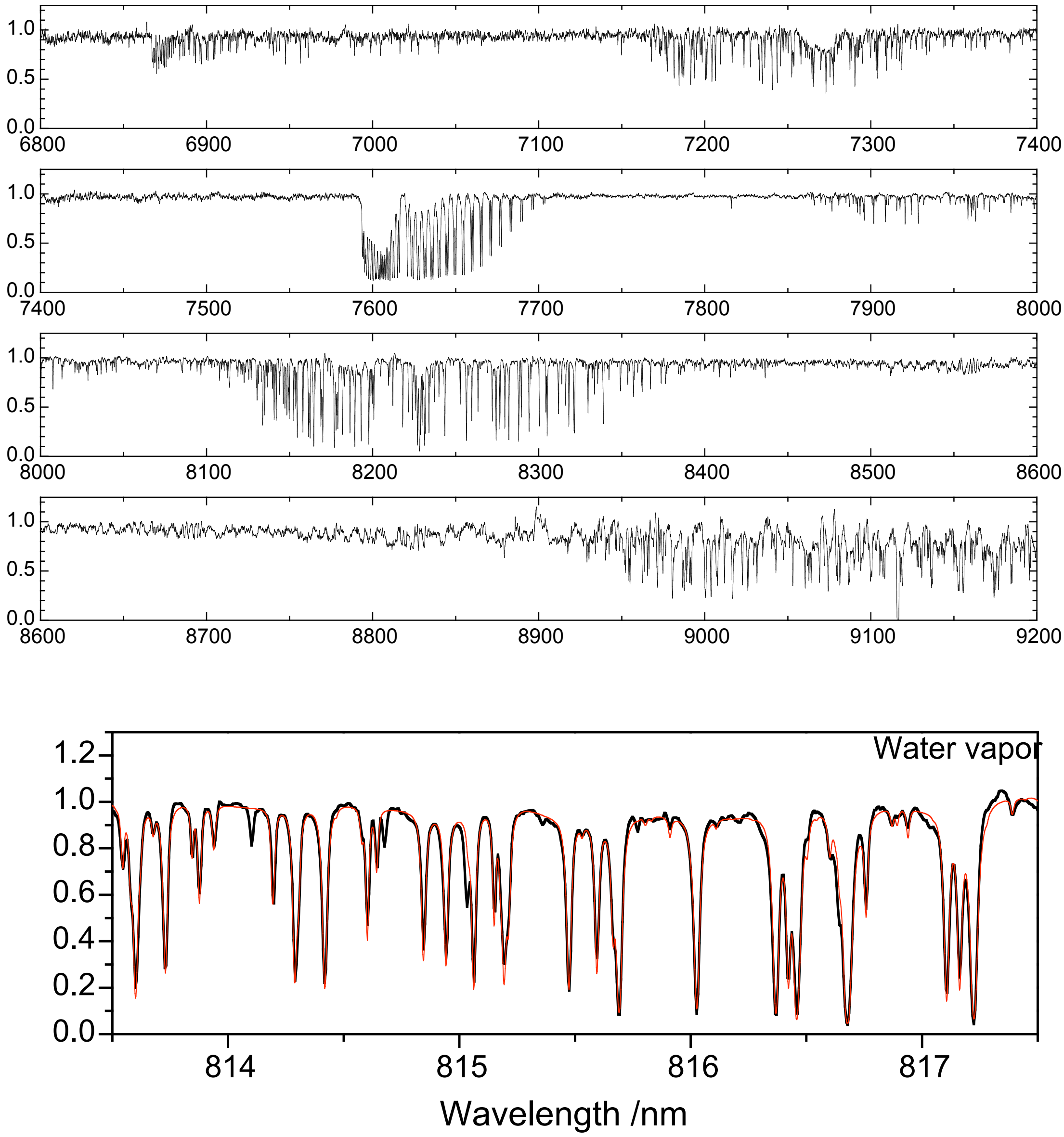


Figure 6. Image of the white-light produced in situ by the Teramobile laser beam (280 mJ, 600 fs with GVD precompensation) with the 2-m Schmidt telescope and a 4-Mpixel CCD. Altitude range: 2.9 – 42 km.

Figure 6 shows a bidimensional image of the white light taken with a CCD in the focal plane of the Schmidt telescope, with a filter opening between 380 and 480 nm. Note that the greyscale is chosen out of 16-bit data to optimize the contrast. The altitude range is 2.9 to 42 km from left to right, with a clear multiple scattering in a cloud layer at 6 km altitude. The efficiency of the white-light generation depends strongly on the chirp. For the same pulse duration, negative chirp (*i.e.* with GVD pre-compensation), the continuum is more intense than with a shorter pulse (150 fs) without precompensation. No supercontinuum is observed when using 600 fs pulses with the opposite chirp. Many

Images have been taken with five different filters, varying the chirp setting and changing between parallel and focused beam.



a)

b)

Figure 7. a) Absorption spectrum from one single exposure (backscatter altitude 4.5 km). b) Comparison of normalized measurement and calculation (after HITRAN) for a part of the H_2O (211)$\leftarrow$ (000) vibration band.

Since many pollutants of interest absorb in the infrared (*e.g.* methane around 1.6 µm, or volatile organic compounds at 3.5 µm), we also investigated the IR domain. During the same campaign we have also, for the first time, performed non-linear lidar measurements of IR light beyond 1.5 µm[19] by connecting fast detectors equipped with broadband filters. These measurements yielded a 10-fold higher signal in the IR than expected from previous laboratory spectra.[13]

We performed spectroscopic measurements with the telescope in the Coudé configuration, coupled with an Échelle spectrograph with a resolution of 0.1 Å. This allowed us for the first time to measure a 240 nm-broad atmospheric transmission spectrum in a single Lidar measurement, up to the bottom of a haze at 4.5 km altitude. (see Figure 7 a). Well-known absorption bands, as the O_2-A band at 762 nm, are visible. Since the oxygen concentration is known, this line can be used to calibrate the temperature of the atmosphere. In the same time, a fit of the water vapor lines with a database such HITRAN[20] (Figure 7 b) yields the absolute humidity. The combination of both values could yield, for the first time, a Lidar measurement of the relative humidity, which is needed for meteorology as well as for climatological models. This example illustrates the power of simultaneous multicomponent remote sensing.

4. AEROSOLS

Aerosols are a key component of the atmosphere. Their characterization is required to estimate their impact on public health, or on the heterogeneous chemistry and the radiative balance of the atmosphere. A further step in the evaluation of new lidar applications lies in the interaction of intense laser pulses with atmospheric aerosols. This nonlinear interaction can provide information about the composition of aerosols. Theoretical studies on this subject have previously been performed.[21] Nonlinear effects induced by ultrashort, high-power laser pulses in aerosol rely either on the microcavity behavior of spherical microdroplets, providing strong feedback for stimulated processes, or on the internal focusing of the incident light, providing high intensity hot spots where the efficiency for nonlinear optical processes is strongly enhanced.

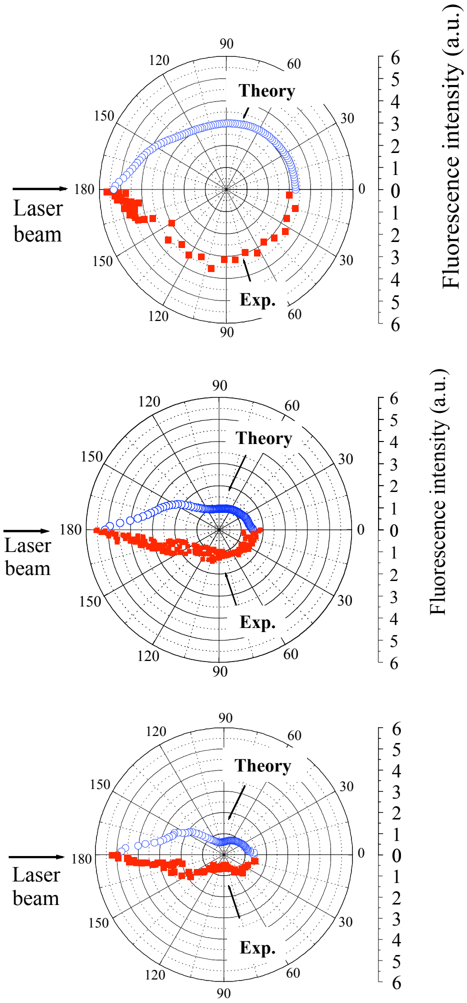


Figure 8. Theoretical and experimental angular distribution of the multiphoton excited fluorescence (MPEF).
Top : 1-PEF, middle: 2-PEF, bottom: 3-PEF.

Such a non-linear behavior is illustrated by the angular distribution of the emission of multiple-photon excited fluorescence (MPEF)[22]. As shown in Figure 8, the 1-PEF is slightly enhanced in the backward direction, and the phenomenon is even stronger for 2- and 3-PEF. This is due to the focusing effect of the droplet, which focuses most of the incident light onto a hot spot (Figure 9), where most of the MPEF will occur. Since the fluorescence emission is isotropic, the emitted light is in turn focused, by reciprocity principle, back to the incident laser direction. This property is highly suitable for Lidar detection, which is based on the collection of backscattered light. Hence, a nonlinear Lidar based on MPEF could bring a powerful analysis capability to range-resolved remote sensing.

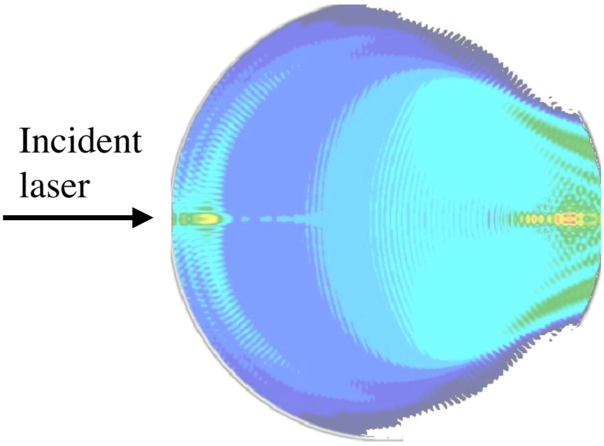


Figure 9. Internal intensity distribution within a transparent sphere illumintated by an incident planar wave.

5. LASER-INDUCED DISCHARGES AND LIGHTNING CONTROL

The possibility to trigger and guide lightning discharges with lasers, in order to get an efficient protection of sensitive installations, has been debated since the early 1970s.[23] Many research groups are working on this subject and have succeeded in triggering and guiding discharges over distances of a few meters, by use of high-energy nanosecond lasers, as well as ultrashort-pulse lasers.[24,25] We have, for the first time to our knowledge, reported guiding and triggering of high-voltage discharges over several meters by laser-induced filaments.[26] High voltage pulses (1.2 µs rise time, 50 µs decay time) of up to 2 MV have been guided over as long as 3.8 m. Figure 10 shows a comparison between discharges without and with laser guiding. For a given electrode gap, the threshold voltage at witch the HV pulse results in a spark was reduced by the presence of the filaments to typically 68% of the free discharge voltage, showing the possibility to trigger the spark before it occurs naturally.

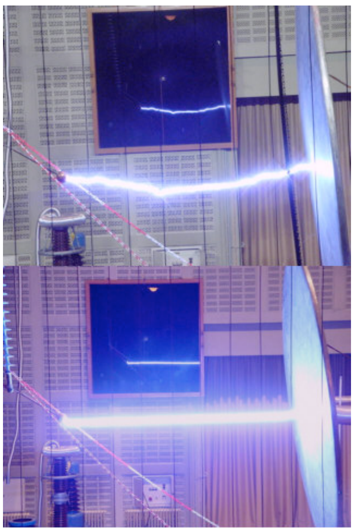


Figure 10. Discharges between a sphere (negative HVelectrode) and a grounded plane. Up: free discharge without laser. Bottom: laser-guided discharge.

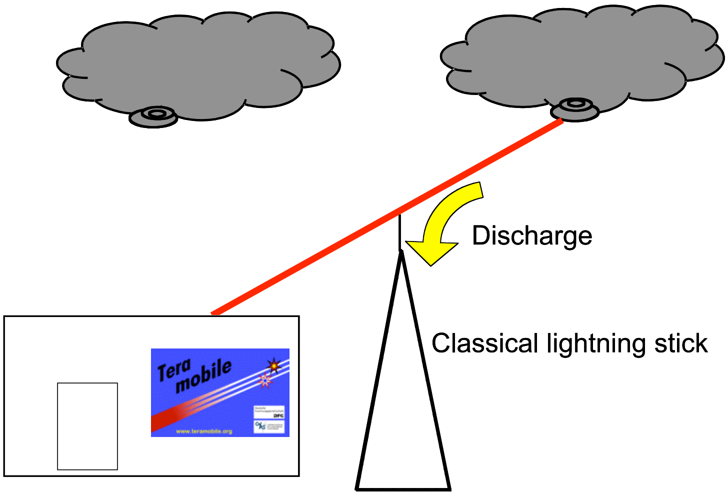


Figure 11. Possible setup of a field experiment to try the control of real atmospheric lightning.

In view of real-scale atmospheric lightning control, a configuration such as that of Figure 11 permits to overcome the limitations due to the limited length of the filaments, as well as to prevent any lightning to strike the laser. In this configuration, the ionized filaments connect a thundercloud to an electrode, *e.g.* a lightning rod, connected to the ground.

6. CONCLUSION

We have presented the current status of the research on the femtosecond lidar, its physical bases and its applications. The construction of the mobile TW laser laboratory, the Teramobile, is a crucial breakthrough to transfer the results from indoor laboratory experiments to field measurements. Recently successful Lidar experiments – of which we have only shown preliminary results – have been performed. Nevertheless, in continuation of the laboratory experiments, field campaigns with the Teramobile to study the basics of the TW laser propagation in air and in aerosol clouds are still needed and planed for the near future. The successful discharge control experiments demonstrate the value of the mobility of the Teramobile system and open the way to real-scale experiments.

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