Femtosecond LIDAR:
new perspectives of atmospheric remote sensing

Miguel Rodriguez*[a], Riad Bourayou[b], Jérôme Kasparian[c], Guillaume Méjean[c], André Mysyrowicz[d], Estelle Salmon[c], Roland Sauerbrey[b], Holger Wille[a], Ludger Wöste[a], Jean-Pierre Wolf[c], Jin Yu[c], Wiebke Zimmer[a]

[a]Institut für Experimentalphysik, Freie Universität Berlin, Germany
[b]Institut für Optik und Quantenelektronik, Friedrich-Schiller-Universität Jena, Germany
[c]Laboratoire de Spectrométrie Ionique et Moléculaire, UMR CNRS 5579, Université Claude Bernard Lyon 1, France
[d]Laboratoire d'Optique Appliquée, ENSTA-École Polytechnique, Palaiseau, France

ABSTRACT

High-power femtosecond laser pulses can lead to strong nonlinear interactions during the propagation through a medium. In air the well known self-guiding effect produces long intense and moderately ionized filaments, in which a broad white-light continuum from the near UV to the mid IR is generated. The forward directed white-light can be used to do range resolved broadband absorption measurements, which opens the way to a real multi-component lidar for the simultaneous detection of several trace gases. On the other hand, enhanced nonlinear scattering and characteristic emission from the filament region, as well as from the interaction of intense pulses with aerosols, can be observed. This opens perspectives towards a novel kind of analysis of atmospheric constituents, based upon nonlinear optics. Additionally, the conductivity of the filaments can be used for lightning control. Here we present the basic concepts of the femtosecond lidar, laboratory experiments and recent results of atmospheric measurements.

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-classic 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 used laser systems are titan-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 Fig. 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.

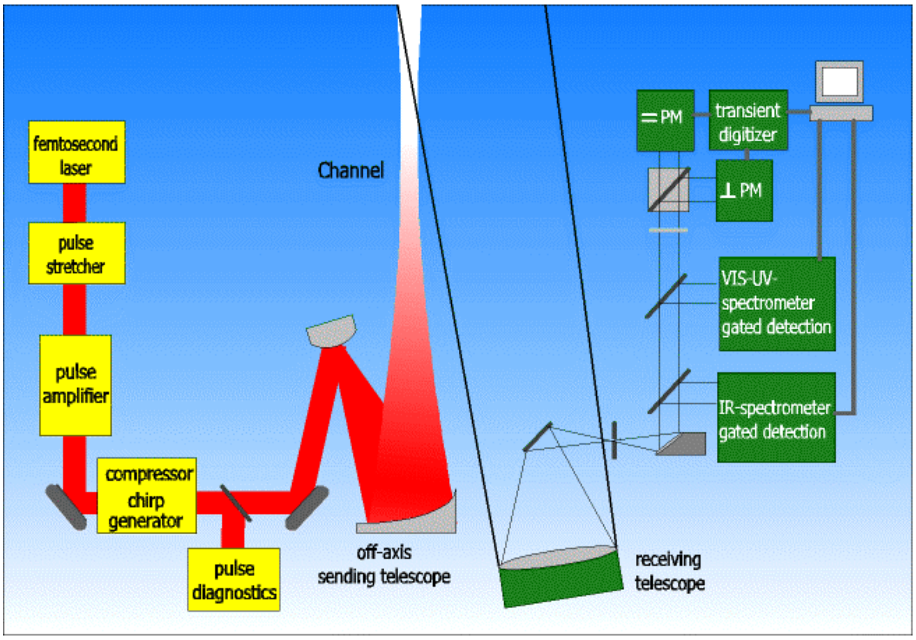
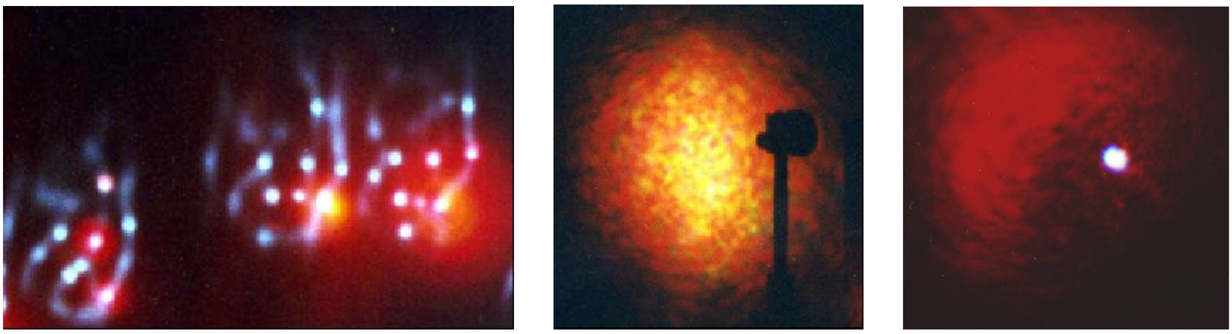


Figure 1: Schematic diagram of the femtosecond lidar setup.

2. NONLINEAR PROPAGATION OF HIGH POWER LASER PULSES IN AIR

At high laser intensities the optical Kerr effect, i.e. the intensity dependent modification of the refractive index, causes self-focussing of the beam. In air the critical power for self-focussing is approximately 2 GW (this is a theoretical value for a diffraction-limited gaussian beam at which the self-focussing length starts to descend from infinity). If no focussing optics is used, this power value is the crucial threshold for the laser pulses to leave the physics of classical light propagation. If the pulse power exceeds this level clearly – in our case by three orders of magnitude – the self-focussing leads to a further enhancement of the intensity and by that to a series of nonlinear optical effects, such as self-phase modulation (SPM) and four-wave mixing (FWM). Those effects, together with the group velocity dispersion

(GVD), lead to a strong modification of the spectral characteristics and the temporal shape of the pulse. Furthermore, when the intensity approaches 10^{13} to 10^{14} W/cm^2 multiphoton ionization (MPI) starts. The appearance of an under-dense plasma results in an intensity dependent reduction of the refractive index, causing a defocusing of 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 filaments. 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 (see Fig. 2 a). 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).[6,9-12] 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.



a) b) c)

Figure 2: Beam profiles of TW pulses propagating through air (real color photographs taken from a white screen). a),b) 100-fs 2-TW pulses after 50 m. a) Multi-filament patterns of three consecutive pulses, diameter of each pattern approx. 5 cm. b) Pattern of a single pulse focussed with an 8-m lens, diameter approx. 20 cm. c) 200-fs 3-TW pulse after 10 m using a 3-m lens. Single filament and red rest light, image diameter approx. 12 cm.

2.1 Control of the filamentation

As one can observe in Fig. 2 a), 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. We have observed quite diverse results using lenses, as can be seen in Fig. 2 b) and c). In some case we see a sparkled pattern of the strongly overlapping emission of many diverging filaments, in others we get a single filament or a narrow bundle of few parallel filaments, with a cone of the red rest light around it. In case of Fig. 2 c) a distortion of the beam profile – which was smoother than in case of a),b) –, e.g. by tilting the lens, leaded to a multi-filament pattern similar to Fig. 2 a). But the filaments become shorter the stronger one focuses the beam.

A different way to control the filamentation distance over ranges beyond a few meters is to use the GVD in air. For that we tune the compressor setting of the CPA chain (see Fig. 1) to give the pulse a linear chirp, i.e. to disperse the spectral parts of the light over the time axis. This makes the pulse longer, but with a so called negative chirp it recompresses during the propagation. Fig. 3 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, while 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 Fig. 3). However, Fig. 3 indicates that the relation between chirped pulse duration and filament distance changes when we go to longer pulses. Long-range experiments, as are planned to be performed with the Teramobile, should show how this relation evolves and what maximal filament distance and length we can achieved.

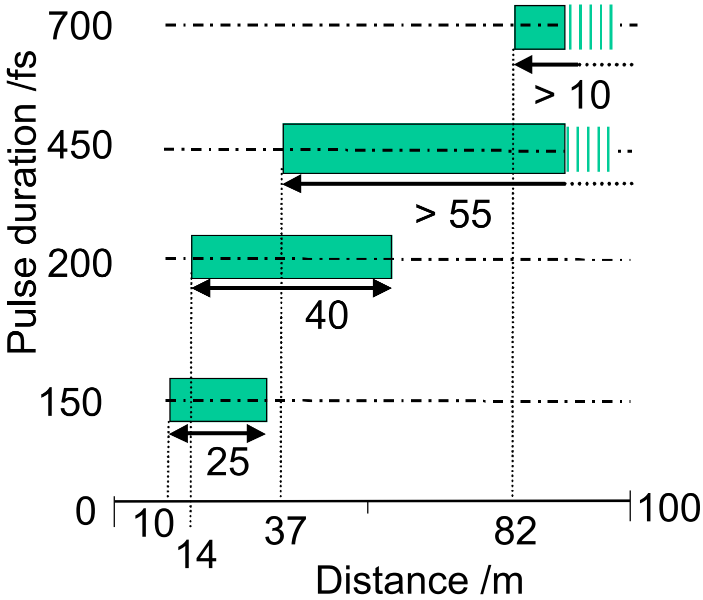


Figure 3: Chirp dependence of the filament position and length. The shortest pulse in the experiment was approx. 100 fs long. Here the value 150 fs stands for a slight positive chirp, the three upper durations represent the length of pulses with negative chirp.

A precise control of the filamentation is very important for the lidar and all other applications that are 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[13] 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 the Section 2.1. The spectral content of the white light has been measured by Kasparian et al.[14] under conditions similar to those of Fig. 2 b), 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 Fig. 4). Spectra in the visible region, similar to the one measured in the laboratory by use of an 10-m lens (see the line plotted in Fig. 4), have been obtained without focussing optics from lidar experiments, i.e. measuring the backscattered light from the atmosphere in 1 km hight.[5] In both cases a dependence of the shape of the spectrum on the pulse power and the chirp setting has been observed. But there is still a lack of knowledge about how the white light varies exactly with the laser parameters. Considering the variety of filament processes, resulting from different laser setups, one can imagine the difficulties to get a complete insight into the dependencies out of experiments. This is especially the case for pure self-focussing if one is restricted to laboratory dimensions. Kilometer-range propagation experiments are planed to be performed with the Teramobile, in order to gain the needed information, e.g. how to obtain the maximal intensity in a certain 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.

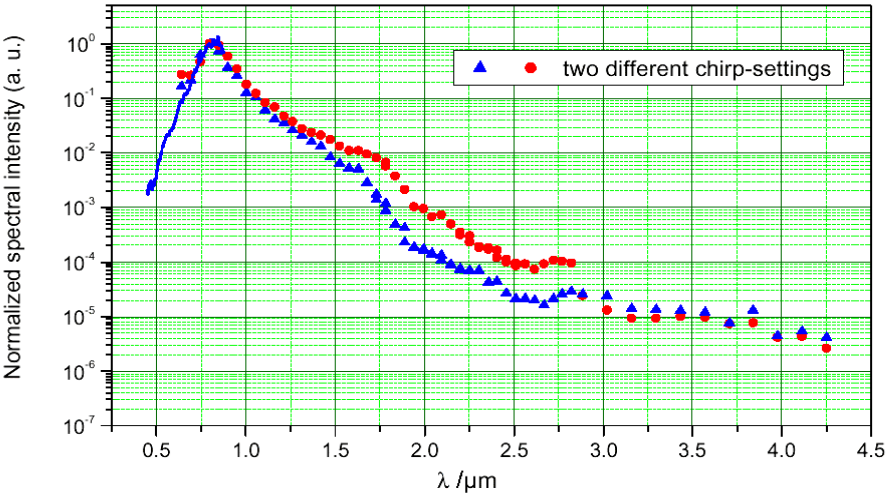


Figure 4: Measured spectra of 2-TW pulses after propagation of 30 m behind a 10-m lens. The triangles represent unchirped pulses, the circles pulses with a negative chirp (both after compressor; the negative chirp mainly compensates the dispersion in the lens).

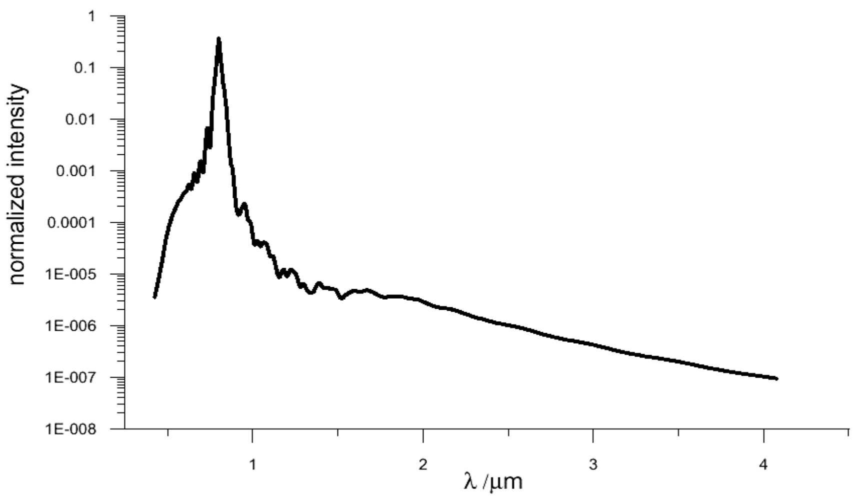


Figure 5: Simulated spectrum of an 85-fs pulse (10 mJ, 800 nm) after free propagation over 1.4 m in air. The radial symmetric calculation starts with a beam diameter of 0.7 mm.[12]

The experimental results have to be compared with numerical simulations. Calculated spectra of the supercontinuum are rare – in contrast to data about the geometry and plasma properties of filaments. Recently Skupin et al.[12] simulated the SCG (Fig. 5) in good qualitative agreement with the experimental results. The model considers the Kerr effect, MPI, GVD and SPM, which turns out to be the main reason for the spectral broadening. Although the pulse energy and propagation range in the simulation are much smaller than in the experiments (both approx. by a factor of 20), a comparison of both is reasonable because in the simulation – which calculates a single filament – the self-focussing length is scaled down by the very small initial beam diameter. The most eye-catching difference is the sharp peak at the fundamental wavelength in the simulation. The flattening of the measured peak could be due to wave-mixing processes that are not considered in the calculation.

3. FEMTOSECOND WHITE-LIGHT LIDAR

The main advantage of lidar over other remote sensing techniques, e.g. differential optical absorption spectroscopy (DOAS),[15] Fourier transform infrared spectroscopy (FTIR)[16] or satellite based spectroscopy,[17] is the high range resolution over large distances, which is achieved by the use of short-pulse lasers (typically a few nanoseconds or less) 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.

The idea of the femtosecond white-light lidar (see Fig. 1) is to combine the advantages of both kinds of remote sensing techniques. Experiments have shown that the white light emitted from self-guided filaments is forward directed and propagating as a collimated beam over large distances.[4,18] In Fig. 2 c) one can see that even when the beam is focussed filaments can form, so that the emitted white light is uncoupled from lens geometry. The Teramobile laser can be sent out in both ways, direct and with a variable focus through an adjustable off-axis telescope. Besides the use of the white-light supercontinuum generated on the first tens of meters behind the laser, a phenomenon measured by Yu et al.[19] could significantly improve non-linear lidar. Due to local laser-induced refractive index gradients, the backward super-continuum emission is significantly enhanced, i.e. more white light is emitted towards the lidar detection system from the filaments than by normal elastic backscattering. This shows again the value of being able to control the filamentation process, as discussed in Section 2.1. The purpose is to be able to place a relatively intense light source at a chosen point in the atmosphere. To achieve this, i.e. to retard the non-linear interaction with the air, the off-axis sending telescope plays an important role because it enlarges the beam diameter to up to 15 cm, which yields a reduction of the initial intensity.

Earlier results of fs white-light lidar experiments have been reported by Rairoux et al.[5] Here we present some preliminary results of very recent measurements with the Teramobile. In a field campaign the Teramobile 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 analysed by a high resolution Echelle spectrograph.

a) 

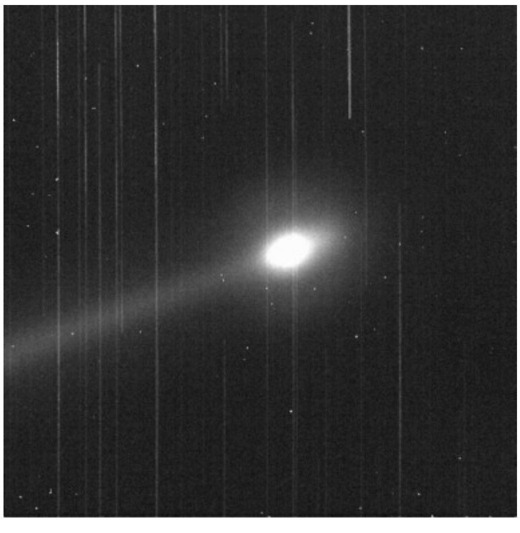
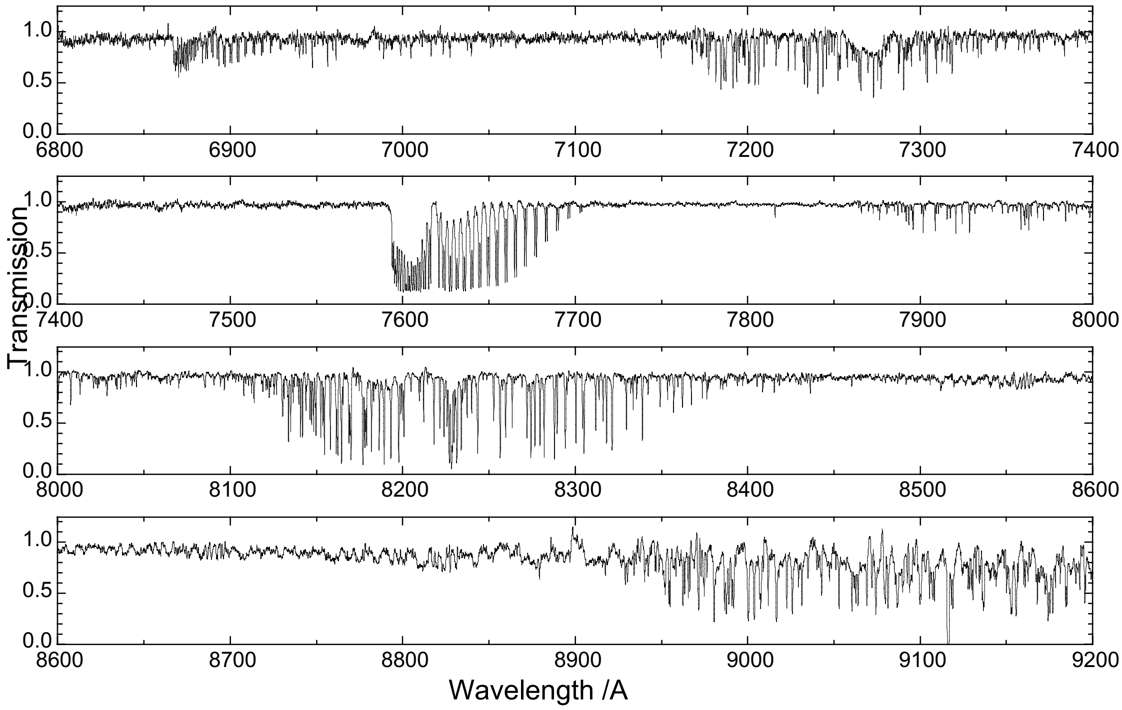
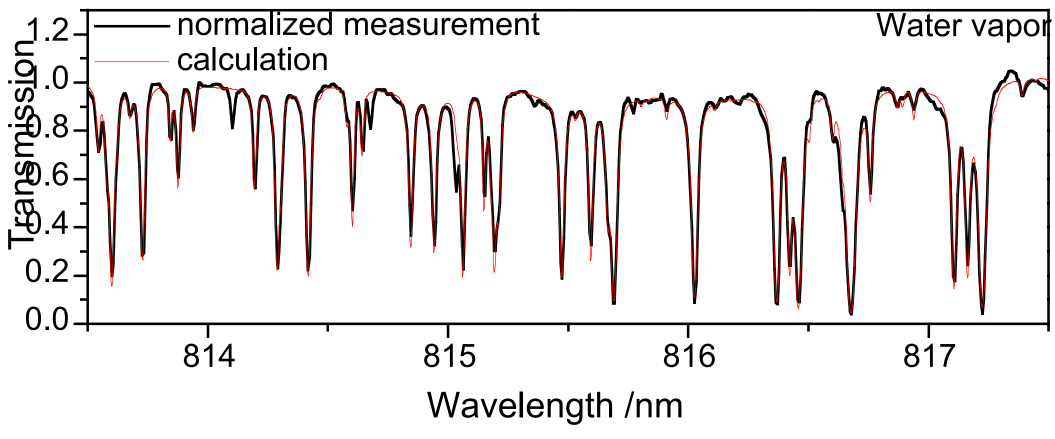
b) 

Figure 6: Images of the Teramobile laser beam (280 mJ, negatively chirped, unfocussed, 10 pulses/s) with a 2-m Schmidt telescope and a 4-Mpixel CCD. a) Pulse duration: 150 fs, filter: Johnson R, exposure time: 5 s, altitude range: 2.8 – 25 km. b) Pulse duration: 600 fs, filter: Johnson B + 2 mm BG 39, exposure time: 360 s, alt. Range: 2.9 – 42 km.

Fig. 6 shows two Schmidt images taken with different color filters (case a: transmission band half-with 560 to 720 nm, 10% at 800 nm, case b: 380 to 480 nm, <10^{-10} at 800 nm). Several pulses are accumulated, according to the exposure time to which the length of the star traces corresponds. Note that the greyscale is chosen out of 16-bit data for each image to optimise the contrast, which can be the criterion for a first comparison of the images. In Fig. 6 a) the beam is visible far beyond a thin cloud layer in an altitude of 9 km. In Fig. 6 b) the Rayleigh scattered light of the blue part of the supercontinuum is visible up to a cloud layer at 6 km. We observed a dependence of the amount of blue light on the chirp setting. With a higher GVD pre-compensation, i.e. a negatively chirped pulse with a duration of 600 fs, we get more intensity than from the 150-fs pulse. Many Images have been taken with five different filters, varying the chirp setting and changing between parallel and focussed 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)←(000) vibration band.

While the Schmidt images contain interesting information about long-range propagation, the spectroscopic measurements performed at the observatory are real examples of the femtosecond white-light application. The Teramobile is equipped with all the needed devices to obtain similar absorption spectra, but here we took advantage of the astronomical installations. Due to the distance of 30 m between laser and telescope the backscatter altitude could be retrieved from the telescope declination, while the lidar detection inside the Teramobile uses the time-to-range relation. The observatory's Echelle spectrograph measures in a wavelength range between 340 and 930 nm in three overlapping channels (e.g. the IR channel 540 – 940) with a resolution of $9 \cdot 10^4$. It is a great advantage to get such a large wavelength range with a single exposure. In Fig. 7 a) we show a 240-nm broad segment of one single measurement of the white light backscattered from an altitude of 4.5 km. Well known absorption bands, as the O_2-A band at 762 nm, are visible. Fig. 7 b) displays a part of the spectrum where the absorption lines belong to water vapor. The calculated spectrum, based on the HITRAN database,[20] fits perfectly to the measured. However, to gain this congruence some parameters, e.g. the exact apparatus broadening, have to be determined. Also the exact baseline of the white light, i.e. the normalization function, has to be fitted simultaneously which is not problematic due to the good match of measurement and calculation. This yields a water vapor concentration of 0.41%, averaged over the propagation path. A series of spectra have been taken from altitudes between 4 and 6 km. Unfortunately the spectrograph did not cover wavelength beyond 1 µm, otherwise the absorption of methane – an important greenhouse gas – could have been measured at 1.6 µm. During the same campaign we also have, for the first time, performed non-linear lidar measurements of IR light around 1.6 µm connecting fast detectors to the telescope, but without a high spectral resolution. These measurements yielded a higher signal in the IR than expected from previous laboratory spectra.[14]

The advantages of the white-light lidar over lidar systems with narrow-band lasers are obvious. The process of retrieval of concentrations is, so to say, auto-calibrating. The narrowness of the absorption lines cause no problem because the analysis is independent of experimental parameters, such as pulse-to-pulse energy fluctuation or the exact laser frequency, which are often difficult to monitor. This aspect becomes crucial when the absorption bands of different species overlap. Especially in the IR it is hard to find absorption lines of trace gases that are totally separated from H_2O and CO_2 lines. And, of course, it is the main goal of the femtosecond white-light lidar research to gain a real multi-component detection technique. The TW laser white-light covers the mid IR where many important pollutants, such as the volatile organic compounds (VOCs) have their absorption bands. The opening of this domain is still difficult, especially on the detector side, and a great challenge for the future.

4. FEMTOSECOND PULSES AND AEROSOLES

A further step in the evaluation of new lidar applications, beyond those described in the previous section, lies in the interaction of intense laser pulses with atmospheric aerosols. In this context the question arises if there are nonlinear optical effects which can provide information about the composition of aerosols. Theoretical studies on this subject have previously been performed.[21] In order to develop a novel lidar technique it is necessary to study first the interaction of femtosecond laser pulses with particles in laboratory experiments. Microdroplets – which represent a major part of the atmospheric aerosol – are attractive systems for the study of several nonlinear optical effects. In microspheres the intensity required for such processes is much lower than in the macroscopic volume due to their strongly curved liquid-air interface. It acts as a lens and focuses the incident radiation onto some small regions inside the droplet. At these areas of high intensity the efficiency for nonlinear optical processes is strongly enhanced. The scattering of a planar electromagnetic wave at an homogeneous spherical particle can be described with the Lorenz-Mie theory.[22] The scattered wave and the internal intensity distribution depend only on the refractive index of the droplet medium and on the size parameter, which is the ratio of the droplet diameter to the wavelength of the incident light. In case of the interaction of femtosecond laser pulses with microdroplets the large spectral bandwidth of the ultra short pulses has to be taken into account. Therefore a large size parameter range has to be considered when calculating the intensity distribution of the incident light inside the droplet by Mie-theory.

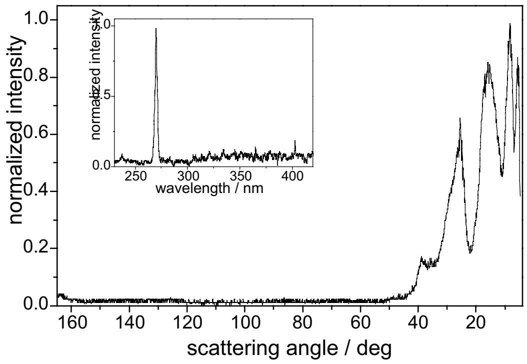


Figure 8: Angular distribution of the THG intensity generated in a water droplet (55 µm diameter) excited with a pump wavelength of 810 nm. Inset: THG spectrum measured under 30°.

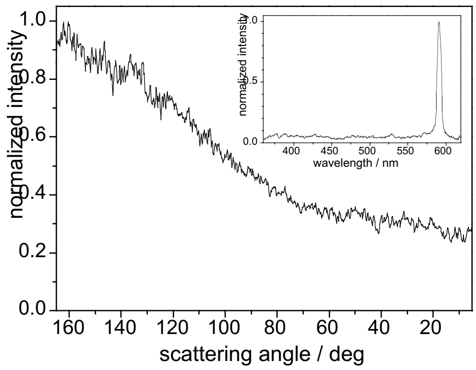


Figure 9: Angular distribution of 589-nm light from a microdroplet containing a 5 molar NaCl solution. The droplet was exited by a focussed laser beam ($4\cdot10^{14}$ W/cm^2). Inset: Spectrum of the plasma emission.

Here we present a few results obtained by use of an 1.5-mJ CPA laser with a 1-kH repetition rate. A piezo-electric nozzle which generates microdroplets in a diameter range of 30 to 50 µm is synchronized with the pulses. At intensities of the order of 10^{11} W/cm^2 third harmonic generation (THG) inside the droplet can be observed. Due to the inversion symmetry of liquids no harmonics of even order can be measured, except for a weak SHG at the surface. In Figure 8 one can see that the angular distribution of the emission is strongly modulated. The structure can be related to the droplet size.[23] At higher intensities – comparable to those inside a filament, here set by moving a focussing lens – plasma is created inside the droplet. Pure water emits a plasma continuum approximately between 300 to 600 nm. But this signal is much weaker than the one that appears at the characteristic wavelength of the sodium line when a NaCl solution is used. The angular distribution of the 589-nm light shows a strong enhancement in backscatter direction (Fig. 9). Hill et al.[24] reported a similarly enhanced backscatter for three-photon fluorescence from droplets. Those results are promising for the remote detection of aerosol compositions with the femtosecond lidar.

5. ADDITIONAL APPLICATIONS

Other atmospheric applications for fs-TW laser systems, in particular for the Teramobile, are based on the ionization induced in the air during the self-channeling process. The filaments act as conducting wires. Numerical simulations and experiments agree that an electron densities of 10^{15} to 10^{16} cm^{-3} is reached inside a filament.[12,25]

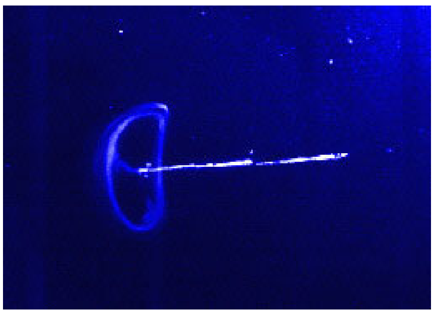


Figure 10: Haze formation in a diffusion cloud chamber along a plasma filament generated with a 1.5-mJ 60-fs laser (the laser pulses propagate from left to right).

5.1 Laser-induced condensation

With the same laser system as used in the droplet experiments (Section 4) we have measured the effect of the ionized filament in humid supersaturated atmosphere. In a diffusion cloud chamber the free charges act as seeds for condensation. In Fig. 10 one can see the condensation strip along the laser filament and some haze in a kind of shock wave from where the filament starts. In this experiment the laser was slightly focussed. In others without a lens a dependency of the self-focussing on the humidity could be observed. Beside the haze production itself, this is an interesting point for the atmospheric propagation.

5.2 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.[26] Many research groups are working on this subject and have succeeded in triggering and guiding of discharges over distances of a few meters, by use of high-energy nanosecond lasers, as well as ultrashort-pulse lasers.[27,28] We have, for the first time to our knowledge, reported guiding and triggering of high-voltage discharges over several metes by laser-induced filaments.[29] Lightning 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. In Fig. 11 one can see a guided discharge (lower image) and in comparison an unguided free discharge (upper image). 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. Because of this reduction we speak of a triggered lightning.

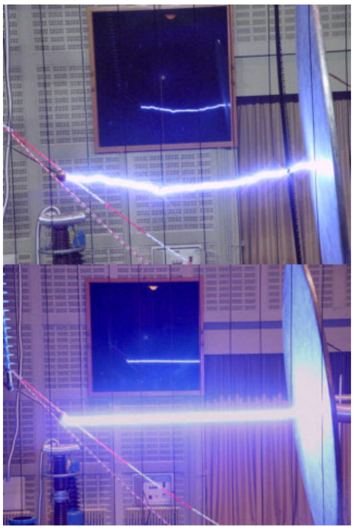


Figure 11: MV discharges between a neg. charged sphere and a grounded plane.
Up: free discharge without laser.
Down: laser-guided discharge.

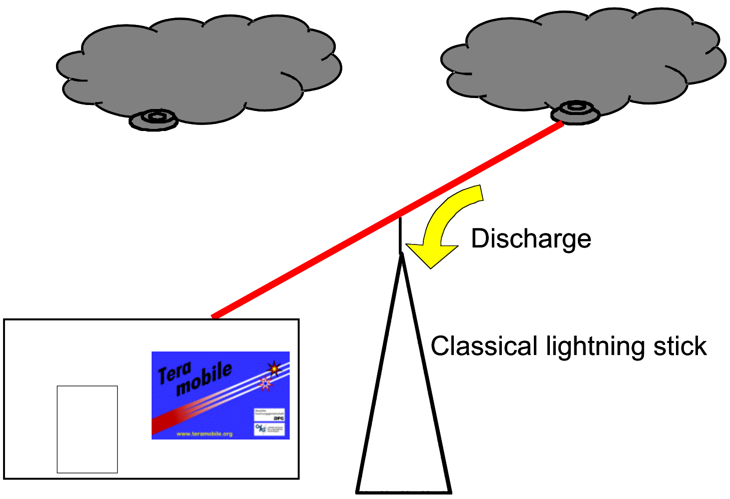


Figure 12: Possible setup of a field experiment to try the control of real atmospheric lightning.

It is not likely that the length of filaments generated by TW-fs lasers will be expandable to such dimensions that they would connect a thundercloud with an electrode, e.g. a lightning rod, on the ground. But, while in a natural lightning, before the final spark, the charged cloud emits streamers "in search of" a discharge path, it should be enough to induce and guide the final jump from a field of streamers in a few hundred of meters height to the ground. For the future, after further preparative laboratory experiments, such a field campaign is planned to be performed with the Teramobile (see Fig. 12).

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 the crucial breakthrough to transfer the results from indoor laboratory experiments to field measurements. Recently successful lidar experiments – of which we have only show 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 are still needed and planed for the near future. On the site of a former military airfield long-range propagation experiments will be possible. Furthermore we plan to continue the studies on the triggering and guiding of high-voltages discharges, with the target to control real atmospheric lightning. The successful experiments performed inside a high-voltage hall demonstrate the value of the mobility of the Teramobile system.

ACKNOWLEDGEMENTS

The authors acknowledge support of the Teramobile project by the Deutsche Forschungsgemeinschaft (Germany) and the Centre National de la Recherche Scientifique (France), as well as sponsoring of travel expenses by the Deutscher Akademischer Austauschdienst (Germany) and the Ministère des Affaires Étrangères (France). J. P. Wolf acknowledges support from the Institut Universitaire de France. We gratefully acknowledge collaboration with and support from the following persons:

- The whole team of the Thüringer Landessternwarte Tautenburg, especially A. Hatzes, H. Lehman, B. Stecklum, A. Scholz, E. Guenther, J. Eislöffel, , J. Winkler, J. Haupt, U. Laux and S. Högner.
- K. Stelmaszczyk from the Institut für Experimentalphysik, Freie Universität Berlin (Tautenburg experiment).
- J.C. Luderer from the Institut für Experimentalphysik, Freie Universität Berlin (cloud chamber experiment).
- T. Fujii from the Central Research Institute of Electric Power Industry, Tokyo (chirp dependency of filamentation and high-voltage experiment).
- W. Kalkner, L. Klingbeil and K. Rethmeier from the Institut für Elektrische Energietechnik, Technische Universität Berlin (high-voltage experiment).

REFERENCES

1. R. M. Measures, *Laser Remote Sensing – Fundamental and Applications*, Wiley Interscience, New York, 1984.
2. D. Strickland, G. Mourou, "Compression of amplified chirped optical pulses," *Opt. Commun*. **56**, pp. 219-221, 1985.
3. S. Backus, C. G. Durfee III, M. M. Murnane, H. C. Kaptetn and H. Nathel, "High Power Ultrafast Lasers," *Rev. Sci. Instrum*. **69**, pp. 1207-1223, 1998.
4. L. Wöste, C. Wedekind, H. Wille, P. Rairoux, B. Stein, S. Nikolov, Ch. Werner, S. Niedermeier, H. Schillinger, R. Sauerbrey, "Femtosecond Atmospheric Lamp," *Laser und Optoelektronik* **29**, pp. 51-53, 1997.
5. P. Rairoux, H. Schillinger, S. Niedermeier, M. Rodriguez, F. Ronneberger, R. Sauerbrey, B. Stein, D. Waite, C. Wedekind, H. Wille, L. Wöste, "Remote sensing of the atmosphere using ultrashort laser pulses," *Appl. Phys. B* **71**, pp. 573-580, 2000.
6. A. Braun, G. Korn, X. Liu, D. Du, J. Squier, and G. Mourou, "Self-channeling of high-peak-power femtosecond laser pulses in air," *Opt. Lett*. **20**, pp. 73-75, 1995.
7. H. Wille, M. Rodriguez, J. Kasparian, D. Mondelain, J. Yu, A. Mysyrowicz, R. Sauerbrey, J.P. Wolf, L. Wöste, "Teramobile: a mobile femtosecond-terawatt laser and detection system," *Eur. Phys. J. AP*, to be published.
8. B. La Fontaine, F. Vidal, Z. Jiang, C. Y. Chien, D. Comtois, A. Desparois, T. W. Johnson, J.-C. Kieffer and H. Pépin, "Filamentation of ultrashort pulse laser beams resulting from their propagation over long distances in air," *Phys. Plasmas* **6**, pp. 1615-1621, 1999.
9. E. T. J. Nibbering, P. F. Curley, G. Grillon, B. S. Prade, M. A. Franco, F. Salin, and A. Mysyrowicz, "Conical emission from self-guided femtosecond pulses in air," *Opt. Lett.* **21**, pp. 62-64, 1996.

10. A. Chiron, B. Lamouroux, R. Lange, J.-F. Ripoche, M. Franco, B. Prade, G. Bonnaud, G. Riazuelo and A. Mysyrowicz, "Numerical simulations of the nonlinear propagation of femtosecond optical pulses in gases," *Eur. Phys. J. D* **6**, pp. 383-396, 1999.
11. J. Kasparian, R. Sauerbrey and S. L. Chin, "The critical laser intensity of self-guided light filaments in air," *Appl. Phys. B* **71**, pp. 877-879, 2000.
12. S. Skupin, U. Peschel and F. Lederer, "Simulation of femtosecond laser pulses propagating in air," *Proc. of the 10th Workshop on Optical Waveguide Theory and Numerical Modelling*, Nottingham, UK, 2002.
S. Skupin, *Simulation der Ausbreitung hochenergetischer Lichtpulse in Gasen*, Diploma Thesis, Fr.-Schiller-Universität Jena, 2001.
13. P. B. Corkum, C. Rolland and T. Srinivasan-Rao, "Supercontinuum generation in gases," *Phys. Rev. Lett*. **57**, pp. 2268-2271, 1986.
14. J. Kasparian, R. Sauerbrey, D. Mondelain, S. Niedermeier, J. Yu, J.-P. Wolf, Y.-B. André, M. Franco, B. Prade, A. Mysyrowicz, S. Tzortzakis, M. Rodriguez, H. Wille and L. Wöste, "Infrared extension of the supercontinuum generated by fs-TW-laser pulses propagating in the atmosphere," *Opt. Lett*. **25**, pp. 1397-1399, 2000.
15. U. Platt, "Differential optical absorption spectroscopy (DOAS)," *Air Monitoring by Spectroscopic Techniques (Chem. Anal., vol. 127)*, M.W. Sigrist (ed.), Wiley -Interscience, 1994.
16. J. Notholt, "The moon as a light source for FTIR measurements of stratospheric trace gases during the polar night. Application for HNO3 in the Arctic," *J. Geophys. Res*. **99**, D2, pp. 3607-3614, 1994.
17. J. P. Burrows, M. Weber, M. Buchwitz, V. V. Rozanov, A. Ladstädter-Weissenmayer, A. Richter, R. de Beek, R. Hoogen, K. Bramstedt, K.-U. Eichmann, M. Eisinger and D. Perner, "The Global Ozone Monitoring Experiment (GOME): Mission Concept and First Scientific Results," *J. Atm. Sci.* **56**, pp. 151-175 1999.
18. S. L. Chin, S. Petit, F. Borne and K. Miyazaki, "The white light supercontinuum is indeed an ultrafast white light laser," *Jap. J. of App. Phys*. **38**, pp. L126-L128 (1999).
19. J. Yu, D. Mondelain, G. Ange, R. Volk, S. Niedermeier, J.-P. Wolf, J. Kasparian and R. Sauerbrey, "Backward supercontinuum emission from a filament generated by ultrashort laser pulses in air," *Opt. Lett*. **26**, pp. 533-535, 2001.
20. L. S. Rothman et al., "The HITRAN molecular spectroscopic database and HAWKS (HITRAN atmospheric workstation)," 1996 edition. *J. Quant. Spectrosc. Radiat. Transfer*. **60**, pp. 665-710, 1998 (see www.hitran.com for HITRAN 2000).
21. J. Kasparian and J.-P. Wolf, "A new transient SRS analysis method of aerosols and application to a nonlinear femtosecond lidar," *Opt. Comm*. **152**, pp 355-360, 1998.
22. J. P. Barton, "Internal and near surface electromagnetic fields for spheroidal particle with arbitrary illumination," *Appl. Opt*. **34**, pp. 5542-5551, 1995.
23. D. Carrol and X. H. Zheng, "Modelling third harmonic generation from microdroplets," *Pure Appl. Opt*. **7**, pp. L49-L55, 1998.
24. S. C. Hill, V. Boutou, J. Yu, S. Ramstein, J.-P. Wolf, Y. Pan, S. Holler and R. K. Chang, "Enhanced Backward-Directed Multi-Photon-Excited Fluorescence from Dielectric Microcavities," *Phys. Rev. Lett*. **85**, pp. 54-57, 2000.
25. S. Tzortzakis, M. A. Franco, Y.-B. André, A. Chiron, B. Lamouroux, B. S. Prade and A. Mysyrowicz, "Formation of a conducting channel in air by self-guided femtosecond laser pulses," *Phys. Rev. E* **60**, pp. R3505-R3507, 1999.
26. L. M. Ball, *Appl. Opt*. **13**, p. 2292, 1974.
27. M. Miki, Y. Aihara and T. Shindo, "Development of long gap discharges guided by a pulsed CO_2 laser," J. Phys. D: Appl. Phys. **26**, pp. 1244-1252, 1993.
28. D. Comptois, C. Y. Chien, A. Desparois, F. Gérin, G. Jarry, T. W. Johnston, J. C. Kieffer, B. L. Fontaine, F. Martin, R. Mawassi, H. Pépin, F. A. M. Rizk, F. Vidal, P. Couture, H. P. Mercure, C. Potvin, A. Bondiou-Clergerie and I. Gallimberti, "Triggering and guiding leader discharges using a plasma channel created by an ultrashort laser," *Appl. Phys. Lett*. **76**, pp. 819-821, 2000.
29. M. Rodriguez, R. Sauerbrey, H. Wille, L. Wöste, T. Fujii, Y.-B. André, A. Mysyrowicz, L. Klingbeil, K. Rethmeier, W. Kalkner, J. Kasparian, E. Salmon, J. Yu and J.-P. Wolf, "Triggering and guiding megavolt discharges by use of laser-induced ionized filaments," *Opt. Lett*. **27**, pp. 772-774, 2002.

* miguel.rodriguez@physik.fu-berlin.de; phone +49-30-838 56119; fax +49-30-838 55567; www.teramobile.org; Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany