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RAPID COMMUNICATION

Remote filament-induced fluorescence spectroscopy from thin clouds of smoke

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Abstract Remote filament-induced fluorescence spectroscopy is used to probe a cloud of smoke, produced from burning mosquito coils, located at a distance of 25 m from the laser source and LIDAR detector. CN, CH and C_2 molecular fragments were identified in the sample. We demonstrate that temporally gated measurement is an efficient technique to easily suppress spectral contaminations, such as white light and atmospheric N_2 fluorescence.

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1 Introduction

In the recent years, due to environmental issues such as global warming and air quality in industrial neighborhoods, there has been an increasing interest towards the development of new tools to control, monitor and quantify atmospheric pollutants species released by the different sources. This worldwide common effort is not only supported by the scientific community. In fact, the countries that supported the Kyoto protocol proposed an emission trading[1] approach that limits the amount of released emissions. A central authority sets a limit on the amount of a pollutant that can be emitted. Companies are issued emission permits and are required to hold an equivalent number of allowances which represent the right to emit a specific amount. Based on this new policy, sooner or later, new tools will be required to efficiently probe and quantify the industries' emissions.

Because of its extraordinary properties (white light generation [1], long plasma channels), filamentation [2] represents an ideal candidate for atmospheric studies [3], such as remote sensing of pollutants [4] and guiding-triggering of electric discharges [5]. This attractive tool is the result of a dynamic interplay between Kerr self focusing induced by the medium's nonlinear intensity dependent refractive index and defocusing by the low density plasma generated through multiphoton/tunnel ionization [6]. In air, this balanced propagation regime stabilizes and limits the light intensity contained in the filament core to an approximate value of 5×10^{13} W/cm^2 [7, 8] which is sufficiently high to ionize any atmospheric molecule. This rather simplified model gives an intuitive view of how a filament behaves. In reality, it is formed from the continuous succession of self-foci arising from the various longitudinal slices of a short pulse [9]. The process starts with the most intense slice at the peak of the pulse and as it focuses, its intensity increases up to a point where it is sufficiently high to ionize the air molecules. The slice is then diffracted out by the generated plasma. Depending on their individual power [10], each slice of the pulse will collapse at a specific position and create a localized plasma. This slice-by-slice self-focusing leads to the formation of a continuous plasma channel aligned along the propagation axis.

Several remote sensing experiments involving filamentation were realized on multiple samples such as gaseous mixtures [11], metallic targets [12, 13], biological dust

[1]Emissions trading. United Nations framework convention on climate change, http://unfccc.int/kyoto_protocol/mechanisms/emissions_trading/items/2731.php.

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grains [14], aqueous aerosol clouds [4, 15, 16] and all of them could be detected several meters away from the source/detector. In this paper, we demonstrate that this remote sensing technique can also work on a distant cloud of smoke. The selected smoke source consisted of mosquito coils from Coghlan's.[2] These repellents, usually made from a cooked mixture of wood flours, binders, colorants, and 0.25% allethrin (the active agent against mosquitoes), have only been selected because of their handiness and have no specific scientific meaning other than generating smoke.

2 Experimental results

A schematic of our experimental setup is presented in Fig. 1. Laser pulses, emitted at a 10 Hz repetition rate by a typical CPA Ti:Sapphire laser system, were focused by a specially designed telescope [17] onto a smoke cloud. The target consisted of a metallic chamber maintained at a negative pressure [4] by an exhaust system evacuating the excess smoke to the outdoor. Eight lighted mosquito coils, uniformly disposed on the chamber's lower internal surface, generated a thin cloud of smoke. The cloud's optical density is characterized, along the filament's propagation axis, by the transmission of a 5 mW He–Ne laser of 7 mm diameter (FWHM). In these conditions, 99% of the light transmitted through the 125 cm thick cloud. The chamber's entrance is placed 25 m away from the focusing telescope in a 30 m long corridor. Beside the telescope, a LIDAR [18] system provided time resolved spectra from the fluorescing target. It consisted of a 25 cm diameter concave mirror of 1.5 m focal length which

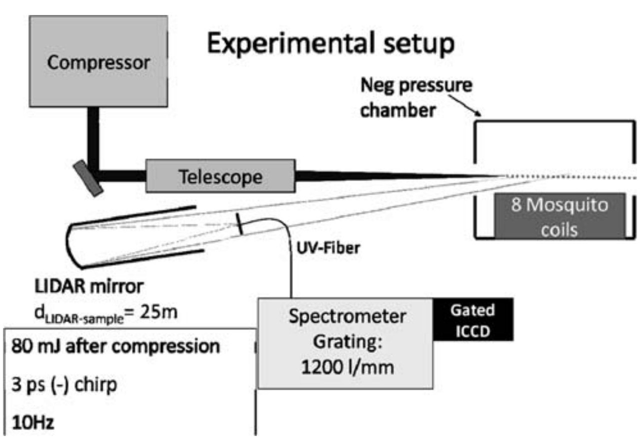

Fig. 1 Short pulses are focused with a telescope on a thin cloud of smoke generated by Coghlan's mosquito coils. The fluorescence signals, resulting from the interaction of the filaments with the cloud of smoke were collected by a LIDAR system and delivered to a spectrometer/ICCD assembly

[2]The outdoor accessory people, http://www.coghlans.com/.

delivered, via an optical fiber bundle, the backscattered light to a spectrometer/ICCD assembly.

Figure 2 shows typical, time-resolved backscattered spectra collected from our smoke target taken at two different times with respect to the pulse's arrival on the sample. The upper trace is measured with a temporal gate of 10 ns starting 3 ns before the pulse hits the target. Therefore, this signal includes the self-transformed (self phase modulation, self steepening, third harmonic generation) white light pulse scattered by the smoke particles and the fluorescence emitted from the recombination of the fragmented molecules. Several molecular transitions were observed, the dominant ones (between 300 nm and 400 nm) originated from fluorescing N_2 and N_2^+ present in ambient air [17]. The large spectral band centered at 540 nm is the second diffraction order of the backscattered third harmonic and does not give any information concerning the sample. Fortunately, this self-transformed white light pulse and its instantaneously created third harmonic can be temporally gated out to unveil the weaker fluorescent transitions masked by the white light.

The second spectrum has been measured with a 10 ns temporal gate width starting 7 ns after the pulse hits the target. The absence of any white light and third harmonic scattered by the cloud significantly decreases the noise level and allows clear detection of the sample's fluorescence. The trace, enlarged in Fig. 3, presents several transitions from the molecular systems $B_2\Sigma$–$X_2\Sigma$ CN, $A_2\Delta$–$X_2\Pi$ CH and the famous $A_3\Pi_g$–$X_2\Pi_u$ C_2 Swan bands. Since they are not observed when the pulse propagates in pure air, these fluorescent molecular fragments originated from the filament's interaction with the smoke cloud's particles.

Besides, because of its short lifetime, the N_2 fluorescence abruptly decays after the interaction while CH, CN and C_2 last much longer. This behavior is exposed in Fig. 4 for CN and N_2 in the spectral window ranging from 377 nm to 395 nm. Each spectrum was measured with a 5 ns temporal gate for different delays after the pulse hit the target. As expected, N_2's much shorter fluorescence decay time allows clear identification of the target's molecular fragments. At the same time, gated measurements become useful not only to eliminate the white pulse scattering from the target, but also to avoid any identification problems encountered due to fluorescing N_2 components overlapping with other systems. This situation is illustrated in Fig. 2 for the CN and N_2 transitions around 357 nm. These two lines are too close to be spectrally resolved (upper curve) by our spectrometer, but they can be temporally resolved (lower curve) to allow the identification of CN.

3 Conclusions

In this paper, we demonstrated that filamentation can be used as an efficient tool to remotely probe thin clouds of

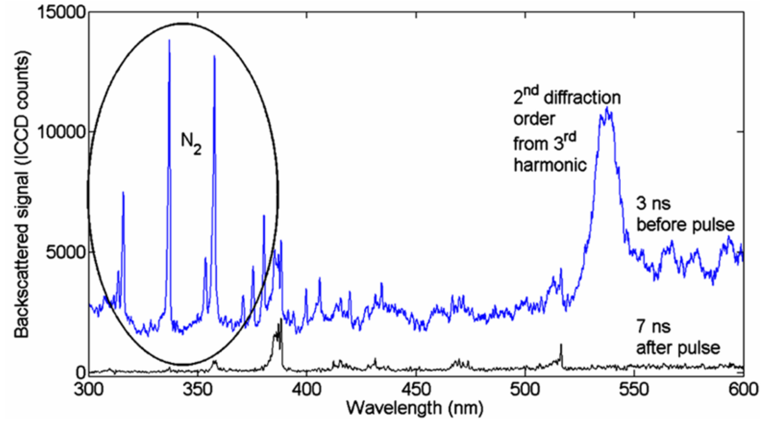

Fig. 2 Gated measurements are required for the identification of the smoke cloud components. In fact, during filamentation, the laser pulse self-transforms into a broadband white light source. This instantaneous light is then scattered by the cloud and collected by the LIDAR. This results in a huge spectral background (*upper curve*) which masks the smoke's fluorescent components (*lower curve*)

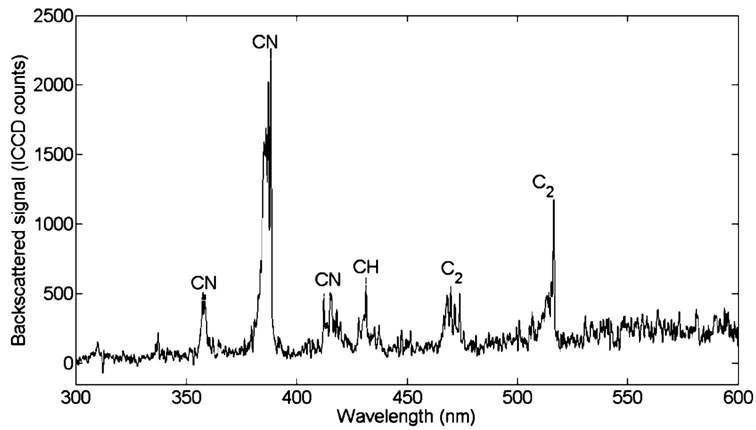

Fig. 3 The lower curve of Fig. 2 is enlarged for easy identification of the spectral components of the fluorescing smoke cloud. Several transitions from the molecular systems $B_2\Sigma–X_2\Sigma$ CN, $A_2\Delta–X_2\Pi$ CH and the famous $A_3\Pi_g–X_2\Pi_u$ C_2 Swan bands are observed

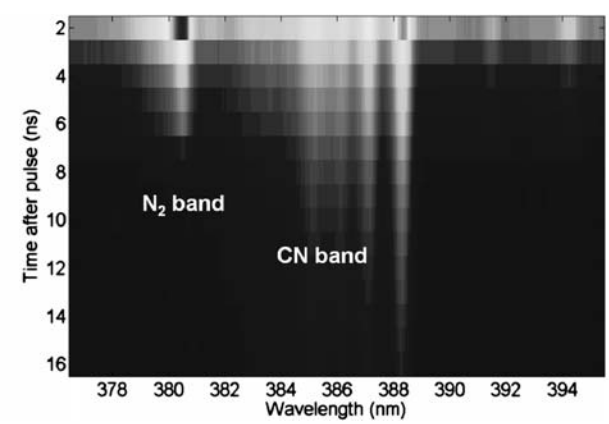

Fig. 4 Time gated measurements of the collected backscattered spectra are presented. The *horizontal scale* presents the spectra's wavelength components and the *vertical scale*, the gate opening time with respect to the laser pulse arrival. This figure illustrates the time evolution of the white light (instantaneous), N_2 and CN

smoke located at a distance. Multiple molecular fragments present in the cloud of smoke, such as CN, CH and C_2, could be identified via the interaction with the intense filaments. The molecules testify that an organic combustion occurred. We demonstrated that the problems linked to the scattering of the self transformed white light pulse onto the target and the spectral contamination of fluorescing atmospheric N_2 molecules overlapping with minor transitions could be avoided with an appropriate temporal gating of the detector.

There are numerous real life applications we could imagine for such a tool. For the militaries, this technique could be used to remotely probe clouds of smoke generated by enemy explosions and identify any toxic or dangerous airborne materials present. Another approach could involve laser systems implemented in wooded areas that could monitor the presence of fires or suspicious clouds of smoke and prevent large scale forests devastations. In a third scenario, the central authority providing the greenhouse emissions permits discussed in introduction, could use mobile laser systems (truck, helicopter, satellite?) to verify that the polluting companies respect their agreements.

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References

1. L. Wöste, C. Wedekind, H. Wille, P. Rairoux, B. Stein, S. Nikolov, C. Werner, S. Niedermeier, F. Ronnenberger, H. Schillinger, R. Sauerbrey, Femtosecond atmospheric lamp. Laser Optoelectron. **29**, 51–53 (1997)
2. A. Braun, G. Korn, X. Liu, D. Du, J. Squier, G. Mourou, Self-channeling of high-peak-power femtosecond laser pulses in air. Opt. Lett. **20**, 73 (1995)
3. J. Kasparian, J.-P. Wolf, Physics and applications of atmospheric nonlinear optics and filamentation. Opt. Express **16**, 466 (2008)
4. J.-F. Daigle, P. Mathieu, G. Roy, J.-R. Simard, S.L. Chin, Multi-constituents detection in contaminated aerosol clouds using remote filament-induced breakdown spectroscopy. Opt. Commun. **278**, 147 (2007)
5. S. Tzortzakis, B. Prade, M. Franco, A. Mysyrowicz, S. Hüller, P. Mora, Femtosecond laser-guided electric discharge in air. Phys. Rev. E **64**, 057401 (2001)
6. S.L. Chin, S.A. Hosseini, W. Liu, Q. Luo, F. Théberge, N. Aközbek, A. Becker, V.P. Kandidov, O.G. Kosareva, H. Schroeder, The propagation of powerful femtosecond laser pulses in optical media: physics, applications, and new challenges. Can. J. Phys. **83**, 863–905 (2005)
7. J. Kasparian, R. Sauerbrey, S.L. Chin, The critical laser intensity of self-guided light filaments in air. Appl. Phys. B **71**, 877 (2000)
8. W. Liu, S. L Chin, Direct measurement of the critical power of femtosecond Ti:sapphire laser pulse in air. Opt. Express **13**, 5750 (2005)
9. A. Brodeur, C.Y. Chien, F.A. Ilkov, S.L. Chin, O.G. Kosareva, V.P. Kandidov, Moving focus in the propagation of powerful ultrashort laser pulses in air. Opt. Lett. **22**, 304–306 (1997)
10. J.H. Marburger, Theory of self focusing. Prog. Quantum Electron **4**, 35 (1975)
11. H.L. Xu, Y. Kamali, C. Marceau, P.T. Simard, W. Liu, J. Bernhardt, G. Méjean, P. Mathieu, G. Roy, J.-R. Simard, S.L. Chin, Simultaneous detection and identification of multigas pollutants using filament-induced nonlinear spectroscopy. Appl. Phys. Lett. **90**, 101–106 (2007)
12. Ph. Rohwetter, K. Stelmaszczyk, L. Wöste, R. Ackermann, G. Méjean, E. Salmon, J. Kasparian, J. Yu, J.-P. Wolf, Filament-induced remote surface ablation for long range LIBS operation. Spectrochim. Acta B **60**, 1025 (2005)
13. W. Liu, H.L. Xu, G. Méjean, Y. Kamali, J.-F. Daigle, A. Azarm, P.T. Simard, P. Mathieu, G. Roy, S.L. Chin, Efficient non-gated remote filament-induced breakdown spectroscopy of metallic sample. Spectrochim. Acta B **62**, 76–81 (2007)
14. H.L. Xu, G. Méjean, W. Liu, Y. Kamali, J.-F. Daigle, A. Azarm, P.T. Simard, P. Mathieu, G. Roy, J.-R. Simard, S.L. Chin, Remote sensing of similar biological materials using femtosecond filament-induced breakdown spectroscopy. Appl. Phys. B **87**, 151 (2007)
15. T. Fujii, N. Goto, M. Miki, T. Nayuki, K. Nemoto, Lidar measurement of constituents of microparticles in air by laser-induced breakdown spectroscopy using femtosecond terawatt laser pulses. Opt. Lett. **31**(23), 3456–3458 (2007)
16. G. Méjean, J. Kasparian, J. Yu, S. Frey, E. Salmon, J.-P. Wolf, Remote detection and identification of biological aerosols using a femtosecond terawatt lidar system. Appl. Phys. B **78**, 535 (2004)
17. W. Liu, F. Théberge, J.-F. Daigle, P.T. Simard, S.M. Sarifi, Y. Kamali, H.L. Xu, S.L. Chin, An efficient control of ultrashort laser filament location in air for the purpose of remote sensing. Appl. Phys. B **85**(1), 55 (2006)
18. R.M. Measures, *Laser remote sensing: fundamentals and applications* (Krieger, Melbourne, 1992). ISBN 0-89464-619-2