# Developments and applications of laser techniques for combustion diagnostics

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## Abstract

In the following presentation a brief review of some past and present achievements in the area of laser diagnostics in combustion environments will be given. Of special interest is the possibility to measure in one laser pulse, ~10 ns, by using a laser sheet and a two-dimensional detector, permitting instantaneous multiple point measurements. This technique has been used for visualization of species, temperatures or velocities. One feature of utmost importance for visualization combustion/flow phenomena is the possibility to make this visualization in real time with adequate time resolution. Most pulsed lasers used for visualization are limited to 10-100 Hz, which is far too low for real turbulent applications. We have therefore for several years developed and applied a special designed laser/detector system for high repetition rate measurements which has been applied for several real-world applications, e.g internal combustion engines. The system can also be used for three dimensional visualization by sweeping the laser pulses through the region of interest and consequent detection with the framing camera. Experiments, mostly using laser-induced fluorescence, from laboratory as well as industrial applications will be exemplified mostly using Laser-Induced Fluorescence.
The presentation will also highlight some other techniques for combustion applications, e.g polarisation spectroscopy, filtered Rayleigh scattering, thermographic phosphorescence and pico-second LIDAR.

### Introduction

A problem but also a challenge with combustion phenomena, is the need for interdisciplinary collaborations to create a thorough understanding of these processes, thus experts in natural sciences, e.g physics, chemistry, mathematics, as well as engineering sciences and combustion devices are needed in order to face real practical combustion problems. During the last decades two major tools have appeared which have given the community new possibilities for detailed studies of combustion processes: new and faster computers which are necessary to model the processes of interest as well as new diagnostic tools for characterization of combustion phenomena and validation of various combustion models.
In the latter field it has been shown that various measuring approaches based on laser techniques have unique properties for diagnostics of combustion processes, see e.g. Ref. 1 and references inthere. The most important features of these techniques are non-intrusiveness in combination with high temporal and spatial resolution. These techniques can be used for measurements of species concentrations, temperatures, velocities and particle parameters (number, sizes, volume fractions).
The present paper will describe some applications of laser spectroscopic techniques for studies of combustion

processes. Since space will neither allow a full coverage of the field nor any basic theory, the paper will be concentrated on certain experimental work in the authors' laboratory. For further details we refer to accompanying references.

The techniques which will be covered are Laser-Induced Fluorescence (LIF), and some emerging techniques, e.g IR polarization spectroscopy, UV filtered Rayleigh scattering and Termographic Phosphorescence. This means that for the many techniques which are not discussed, e.g. Raman scattering, CARS, DFWM, LII e.t.c., we refer to Refs. 1-4.

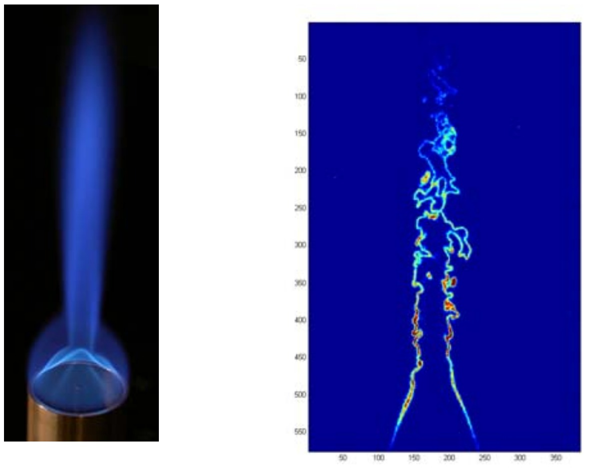
## Laser-Induced Fluorescence, LIF

The laser technique which probably has received the largest attention for combustion diagnostics is Laser-Induced Fluorescence, LIF. In this technique the laser beam is tuned to an atomic or molecular absorption transition and by detecting the atomic/molecular fluorescence emission, it is in principle possible to infer both species concentrations, temperatures and velocities. The main advantages with this technique is that it is very sensitive, sub ppm levels can be measured, and that, as described below, two-dimensional measurements can be performed. It is mainly radicals of importance in combustion which are measured, e.g. OH, $C_2$, CH, NO excited by a one photon resonance. However, species absorbing in the deep vacuum ultraviolet spectral region can also be measured using two-photon resonances, e.g atoms like O, H, N, C and CO, $NH_3$. For a thorough description of the LIF technique we refer to Refs. 1, 2.

### 2D LIF developments and applications

The LIF technique has become a very important tool in characterization of combustion processes as shown below, since it gives a unique possibility, in time and space, to visualize combustion processes in two dimensions *in-situ*. An example of a recent 2D single shot application using LIF is shown in Fig 1. Here the CH radicals were visualized in a jet flame using a powerful Alexandrite laser [5]. In this case the Alexandrite laser, tunable in the near IR spectral region, was frequency doubled to the spectral region where the CH radical absorb, ~387 nm. In this case, since the laser pulse is rather long, ~100 ns, the linewidth can be made rather broad and the laser energy is high, ~50 mJ/pulse, a detectivity about two orders of magnitude higher than what has been reported before could be achieved.

A natural development of the one-species, single shot 2D LIF technique visualization is the extension to visualization of two or more species. This can be done in different ways and in Fig. 2a is shown a two-laser/two detector set-up for simultaneous species visualization in an engine [6]. In this case one laser system was tuned to the OH absorption around 283 nm with subsequent fluorescence around 310 nm, the other laser system is a frequency tripled YAG laser at 355 nm which by coincidence has a spectral overlap with absorption lines of formaldehyde with fluorescence emission in the visible region



a b

*Figure 1. Photograph of a turbulent jet flame (left) and the corresponding 2D LIF image of CH radicals in the flame (right)*

As can be seen in Fig. 2b, which are experiments from a direct injected HCCI engine, it is possible to visualize both species at different crank angle degrees (still the images are recorded from different cycles).

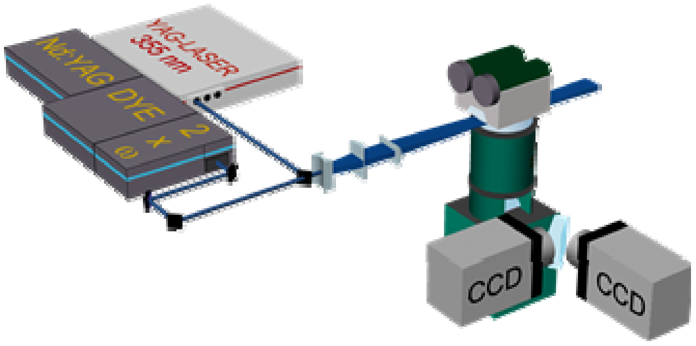


*Figure 2a. Experimental set up for dual species visualization in an engine*

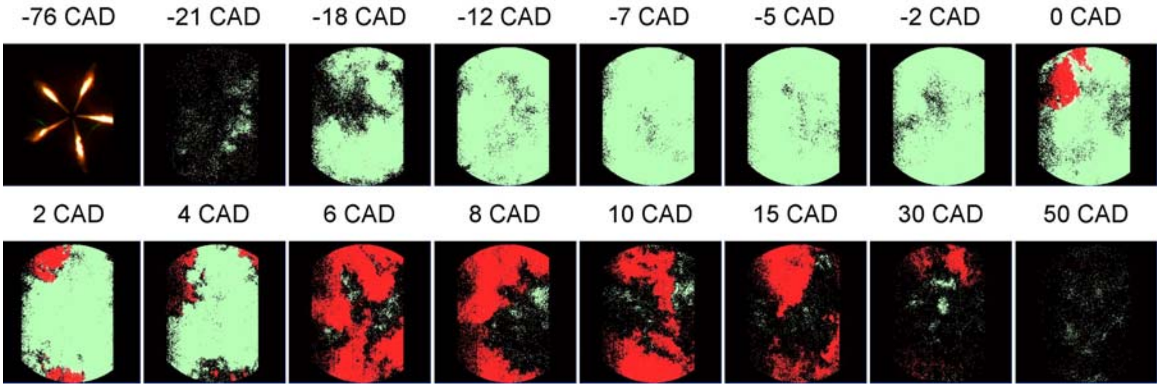


*Figure 2b. Simultaneous images of formaldehyde (green) and OH (red) at different crank angle degrees from a direct injected HCCI engine*

### Temporally resolved LIF visualization

Until the late nineties almost all measurements and experiments reported on were made by more or less standard high power Nd:YAG/Excimer lasers. One problem with these pulsed laser is the repetition rate, which is limited to ~10-100 Hz, and thus clearly too slow to follw fast turbulent events, e.g. inside an engine. We have for amost a decade been working with the development of a laser system for high peed visualiation [7]. The system is capable of producing a rapid sequence of up to 8 laser pulses with a temporal resolution ranging from microseconds to milliseconds. The system consists of a Nd:YAG laser cluster and a high speed camera. The laser source of the high speed laser diagnostic system is a cluster consisting of 4 standard flashlamp pumped Nd:YAG lasers. Each laser in the cluster consists of a Q-switched Nd:YAG oscillator and a single amplifier, emitting laser pulses with a duration of 7 ns.

A single fourth harmonic generation crystal, converting the 532 nm pulse train to 266 nm can optionally be inserted near the output of the unit. The four individual lasers can be fired in series with time delays ranging from 0 up to 100 ms, where the upper limit is given by the overall repetition frequency of the system (10 Hz). By switching the Q-switch twice during the flashlamp pulse duration, each of the Nd:YAG lasers can be fired two times with a short separation between pulses. By interleaving the double pulses from the four lasers the time separation between pulses can be reduced down to 6.25 μs.

Single-cycle resolved measurements using the high-speed laser and detector system, described above, have been performed on different types of flames and for investigating how combustion proceeds in various engines, see e.g [8]. Results of this sort can hopefully lead to a better understanding of engine design and to future improvements in engine efficiency engines with a corresponding reduction in pollution. In Fig 3 is shown single-cycle resolved fuel visualization measurements performed by means of PLIF with the use of an excitation wavelength of 266 nm.

The results of this engine application show that it is feasible to perform single-cycle-resolved diagnostics in engines by use of the high-speed detection system.

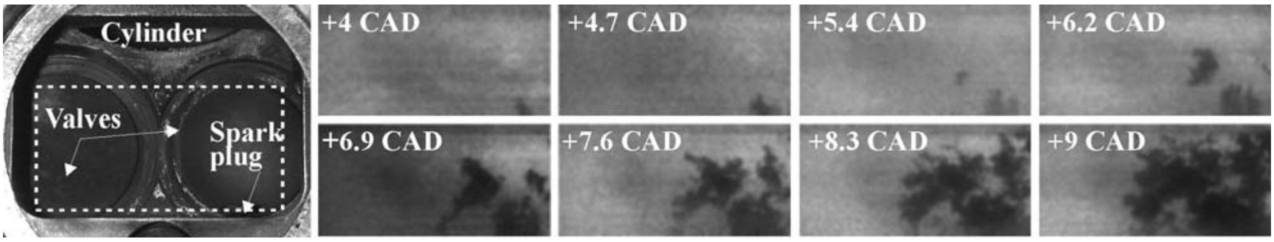


*Figure 3. A fuel tracer LIF sequence in the SI-engine showing the engine's fuel consumption. The dark regions correspond to burnt areas in which the fuel has been consumed, whereas in the bright regions combustion has not yet started. The region involved, 50×25 mm in size, is shown to the left.*

### 3D measurements

So far imaging has been described in two dimensions. Since combustion is a phenomenon that occurs in all spatial directions, instantaneous imaging in three dimensions provides interesting and useful information about the combustion process. When two-dimensional imaging is performed, the features of the flow that become evident depend upon the orientation of the laser sheet. A single slice through the flow does not contain all of the information required to fully describe the structure of the flame. Three-dimensional visualization is feasible by using the ultrafast laser/detector system by sweeping the laser beam through a flow or flame using a rapidly rotating mirror [9]. It is then possible to record stacks of eight closely spaced planar images. The experimental set-up is illustrated in Fig. 4.

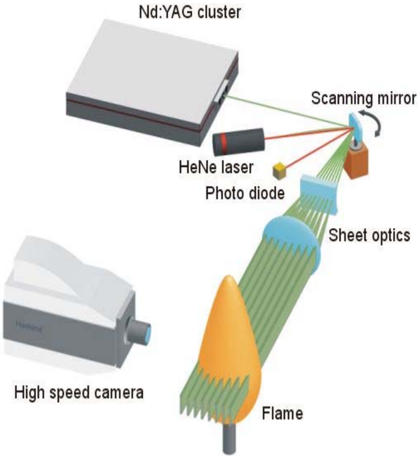


*Figure 4. Experimental set-up for 3D LIF imaging*

A cylindrical and a spherical lens are used to form light sheets, by positioning the scanning mirror at the focal point of the spherical lens the sheets became parallel. The separation between the laser sheets determines the smallest scale that can be resolved, and is thus determined by the expected length scales of the flow. Normally the separations between the laser sheets range from 0.5 to 1 mm. The total acquisition time of the 3-D data, around 70 μs, is sufficiently short to freeze the low Reynolds number flows studied, which can be checked by recording image series without scanning the mirror. However, as short acquisition times as possible are generally desired, to freeze also higher Reynolds number flows. With the system presented here, however, the mirror speed limits the minimum acquisition time, for the desired sheet spacing. The minimum acquisition time, for an eight image measurement, is also limited by the shortest double pulse separation possible (6.25 μs). For proper images it is of course important that the depth of field of the imaging optics exceeded the dimensions of the measurement volume.

In Figure 5 is shown an example where three dimensional measurements of formaldehyde have been performed in a DME/air flame [10].

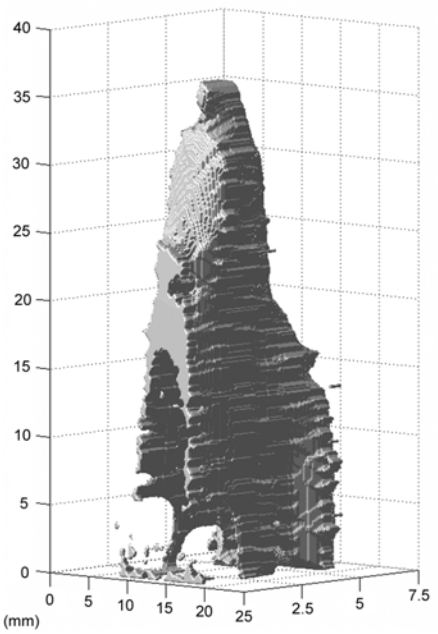


*Figure 5. Single shot 3D image of formaldehyde in a flame*

### **“New” techniques**

Since LIF is a non-coherent laser technique, meaning that the emission is scattered in all directions, it would be very attractive to have a sensitive coherent technique where the signal is generated as a new laser beam, permitting measurements in an environment with limited optical access. Two techniques, yielding the possibilities for minor species detection in multiple points and where the signal is generated as a new laser beam, are Degenerate Four-Wave Mixing, DFWM, and Polarization Spectroscopy, PS. For the former technique see Refs. 1, and 3 for further details.

Polarization Spectroscopy (PS) is a technique from the late seventies, mainly used for Doppler free experiments [11], which has been shown to have a great potential for combustion diagnostics. Very briefly, in this technique, which is based on saturation spectroscopy, a strong well-polarized laser beam tuned to a molecular or atomic transition induces an optical anisotropy of the molecules by optical pumping among the magnetic sublevels of the specific transition probed. The optical anisotropy may be detected by crossing the pump beam by a weak probe beam between two crossed polarizers. In the absence of the pump beam only leakage through the second polarizer will be detected. When the pump beam is present the anisotropy will change the state of polarization and a signal will be detected through the second polarizer. This technique will thus also give the signal as a coherent beam and has a detection limit that is almost comparable with LIF. The application of PS for combustion diagnostics was demonstrated in the mid eighties and in the nineties the technique found new applications and the first demonstration of two dimensional visualization was made in 1993 [12]. In this experiment OH was probed by using a sheet shaped pump laser in combination with an unfocused probe beam.

The PS technique has during the last years also been extended to measurments of species absorbing in the IR part of the spectrum, which gives unique possibilities for spatially and temorally resolved experiments, e.g of individual hydrocarbons, CO, CO2 and H2O, see Refs. 13,14. In Fig. 6 is given an example where methane and ethan have been simultaneoulse measured and spectrally identified using IRPS around 3 µm.

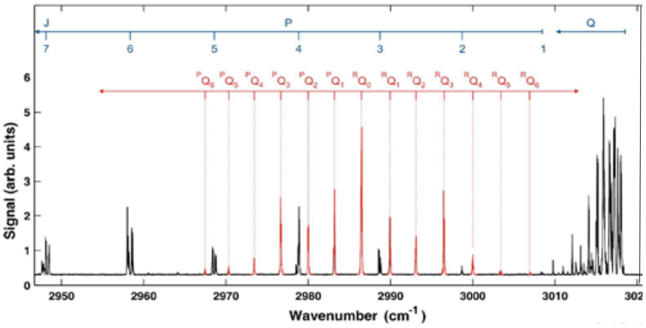


*Figure 6. IRPS spectra of $CH_4$ and $C_2H_6$*

Another emerging technique for thermometry in combustion environments is the use of Thermographic Phosphors, TP. Thermometry based on TP utilises the physical properties of the phosphor particles for assessing temperature. The phosphor particles used for thermometry are usually inorganic particles some 1 - 5 µm in diameter. Such a phosphor consists of a host material and a doping agent from which the light is emitted. A large number of different phosphors are produced today. These cover a wide range of temperatures, from cryogenic temperatures up to 1700 C or higher, making them suitable for many different applications, see Ref. [15] for a review of the technique and various applications. So far the technique has mostly been used for measurments on surfaces. Once deposited on the surface of interest and excited by a suitable wavelength, mainly UV light, the phosphor particles emit an intense luminescent light having a duration on the order of $10^{-7}$ s. After excitation of the thermographic phosphor, the subsequent emission is imaged onto a detector. The temperature can then be deduced from the spectral or temporal properties of the recorded signal. This technique provides a high quantum yield, two-dimensional measurements and remote thermometry, as well as a high degree of accuracy. In contrast to pyrometry, it is not influenced by the emissivity of the material, background reflection from the surroundings or light absorption by optical windows or by surrounding gases. These advantages have allowed thermographic phosphors to be used in a wide variety of applications and in harsh environments, e.g. fire applications [16] and in engines [17]. The technique has also been developed also for spray studies [18] as well as gas thermometry [19]. Very recently new blue emitting phosphors with short lifetimes have been investigated and applied for high precision measurements in burning droplets, see Fig. 7.

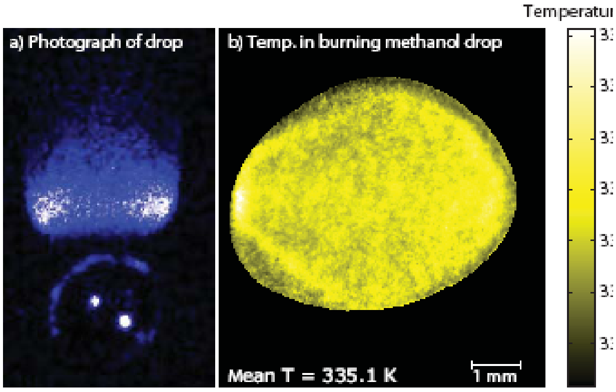


*Figure 7. Burning droplet (left,) single shot 2D temperature measurement in the droplet (right)*

Other techniques which we may see emerge for potential practical applications are single-ended measurements based on a picosecond LIDAR approach [20], Filtered Rayleigh scattering [21 ] and techniques for proper evaluation of measurements in dense sprays avoiding multiple scattering, Ballistic imaging and more recently Structural planar illumination, SLIPI [22].

## Conclusions

As have been demonstrated during the last decade and exemplified in this paper, various laser spectroscopic techniques have become of great importance in characterizing various combustion processes. It is strongly believed that fundamental studies of existing as well as new techniques and their subsequent application, e.g. for characterization and optimization of combustion apparatus, are of crucial importance. It is also believed that the future will show an increased collaboration between diagnosticians and modellers and that these activities will be made in close collaboration with relevant industry, so that cars, trucks, aeroplanes, boilers etc of the next generation are further optimized in terms of fuel efficiency and pollutant formation.

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