

## **Investigation of In-Cylinder Soot Formation and Oxidation by Means of Two-Dimensional Laser-Induced Incandescence (LII)**

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### **Abstract**

Increasing requirements on the environmental acceptability of internal combustion engines led to strong efforts to meet the continuously getting stricter emission regulations. New concepts of combustion and injection have been developed together with appropriate techniques of exhaust gas aftertreatment. For diesel engines especially modern high pressure injection systems, such as the application of distributor pump and common rail have been applied successfully. However, internal mixture formation, ignition and combustion for this new systems are not completely understood so far. Optical measurement techniques offer the possibility of the in-situ investigation of the combustion process. An appropriate method for soot measurements, which is without source of interference also applicable to technical systems, is given by laser-induced incandescence (LII). The principle of this technique is to heat up the soot particles by a highly energetic laser pulse to temperatures well above ambient temperature and to detect the resulting thermal radiation. For appropriate experimental conditions, the signal can be shown to be proportional to the soot mass concentration within the detection volume.

In this work, basic features of the technique are discussed in detail, especially with respect to the application within diesel engines. One aspect of particular interest is the range of detection wavelengths, which has to ensure a mass proportional signal of sufficient magnitude, but also the suppression of flame luminosity and especially elastic scattering of walls, droplets and dust. Another important point to consider is attenuation of both the incident beam and the signal due to light absorption, scattering and blinding of the optical access. Measures to minimize these influences are discussed.

The soot formation and oxidation process inside the combustion chamber of a DI diesel engine was investigated by means of two-dimensional LII. For this purpose, a thin light sheet was introduced into the piston bowl of an optically accessible diesel engine, which is very close to the serial standard and driven with standard diesel fuel. The detection was performed perpendicularly to the incident beam through a transparent piston window and a mirror which was mounted inside the slit of the elongated piston. First results of a common rail injection system are presented for a mini-sac-hole nozzle. Sequences of the LII signal and flame luminosity were measured in a time interval from before top-dead-center (TDC) to about 30 degrees crank angle after TDC. The comparison to investigations of natural flame luminosity, which can be detected simultaneously, is shown to give valuable additional information. It is, e.g., possible to detect a LII signal without the simultaneous occurrence of flame luminosity, which can be interpreted as originating from soot, which will not be further oxidized due to the end of the diffusion flame. This portion will be emitted later on within the exhaust stroke. Generally, the flame does not cover the complete piston bowl for the operation conditions investigated in this study.

## Introduction

The development of new combustion concepts, which is enforced by continuously stricter demands on the environmental acceptability of internal combustion engines, heavily relies on an improved knowledge of internal combustion phenomena. High pressure common rail systems, which offer additional flexibility of injection parameters connected with improved mixture formation, are increasingly applied to modern diesel engines in order to lower both pollutant emission and fuel consumption. However, the mechanisms of spray propagation, mixture formation, ignition and combustion are not completely understood up to now.

The application of optical measurement techniques as an extension of conventional techniques allows a deeper insight into these basic mechanisms. Investigations can be performed with high spatial and temporal resolution and without disturbance of the in-cylinder flow and combustion.

Regarding soot formation and oxidation laser-induced incandescence (LII) has been shown to be an appropriate tool even for technical systems. This technique offers the possibility of the quantification of soot mass concentration and can be performed to obtain two-dimensional information within a given detection plane. It has been shown to give reliable results within laboratory flames by many researchers, e.g. by Shaddix et. al. [1994] and Quay et. al. [1994]. Even some in-cylinder measurements have been published, e.g. by Dec [1992] and Inagaki et. al. [1999], within the last few years, which, however, rely on considerable modifications of the engine or the application of substitute fuels. Recently, the LII technique was firstly applied to exhaust gas measurements by Schraml et. al. [1999, 2000] and the determination of additional quantities, like the soot primary particle size or the agglomerate size was added to gain additional information concerning soot morphology by Will et. al. [1995, 1996, 1998].

In this work, LII is successfully applied to a research engine, which is very close to the series standard and is driven with standard diesel fuel. The results show almost no interference with reflections from walls and droplets and demonstrate the usefulness of this technique.

## Theory

Basic principle of the laser-induced incandescence (LII) technique is to heat up the soot particles within the probe volume to about vaporization temperature by means of a highly energetic laser pulse and to detect the strongly enhanced thermal radiation with an appropriate detector. The relevant mechanisms of power absorption and heat loss have to be included into a power balance to calculate the resulting particle temperature  $T$  after laser irradiation. The differential equation reads [Will et. al., 1998]

$$Q_{abs} \frac{\rho d_p^2}{4} E_i - \Lambda (T - T_0) \rho d_p^2 + \frac{\Delta H_v}{M} \cdot \frac{dm}{dt} - \rho d_p^2 \int \mathbf{e}(d_p, \mathbf{I}) M_1^b(T, \mathbf{I}) d\mathbf{I} - \frac{\rho d_p^3}{6} \mathbf{r} C \frac{dT}{dt} = 0, \quad (1)$$

and generally has to be solved numerically. The individual terms denote in the given order laser absorption (absorption efficiency  $Q_{abs}$ , laser irradiance  $E_i$ ), heat transfer to the surrounding medium (heat transfer coefficient  $\Lambda$ , ambient temperature  $T_0$ ), heat loss due to vaporization (heat of vaporization  $\Delta H_v$ , molar mass of carbon  $M$ , mass loss  $dm/dt$ ), thermal radiation (emission efficiency  $\mathbf{e}$ , blackbody spectral radiant exitance  $M_1^b$ , wavelength  $\mathbf{I}$ ) and change of internal energy (density  $\mathbf{r}$ , specific heat of solid carbon  $C$ ). The individual terms are discussed in more detail by Melton [1984], Dasch [1984], Hofeldt [1993] and Will et. al. [1998].

In setting up equation 1 spherical primary particles of diameter  $d_p$  are assumed, which are allowed to build agglomerates with point contact and which do not inhibit heat release of one another. This assumption is generally fulfilled by soot agglomerates consisting of chain-like structures with fractal dimensions of 1.5-1.8 [Puri et. al., 1993]. From the solution of equation 1 the relative LII signal

$$S_{LII} \propto d_p^2 \int R(\mathbf{I}) \mathbf{e}(\mathbf{I}, d_p) M_1^b(T, \mathbf{I}) d\mathbf{I} \quad (2)$$

follows from the integration of the spectral radiant exitance, given by

$$M_1(T, \mathbf{I}) = \mathbf{e}(d_p, \mathbf{I}) \cdot \frac{2\rho h c^2}{I^5} \cdot \frac{1}{\exp\left(\frac{hc}{IkT}\right) - 1}, \quad (3)$$

weighted by the spectral characteristics  $R(\lambda)$  of the detection path.

*Soot Mass concentration*

From the solution of the power balance it can be observed that temperatures of well above 4000 K are reached (Fig. 1) if high values of the laser irradiance are chosen.

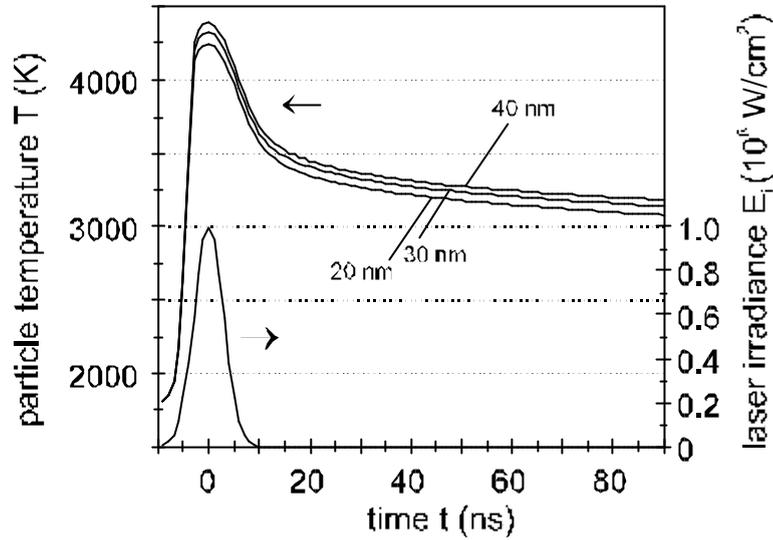


Fig 1.: Temperature of differently sized soot particles after laser heating.

At this moment which nearly coincides with the maximum of laser irradiance, the power balance (equation 1) can be simplified to

$$Q_{abs} \frac{\rho d_p^2}{4} E_i^{max} + \frac{\Delta H_v}{M} \cdot \frac{dm}{dt} = 0, \quad (4)$$

as all other heat loss mechanisms are in very good approximation negligible compared to vaporization at these temperature. This can be clearly seen from figure 2 which gives the relative magnitude of the individual heat loss mechanisms.

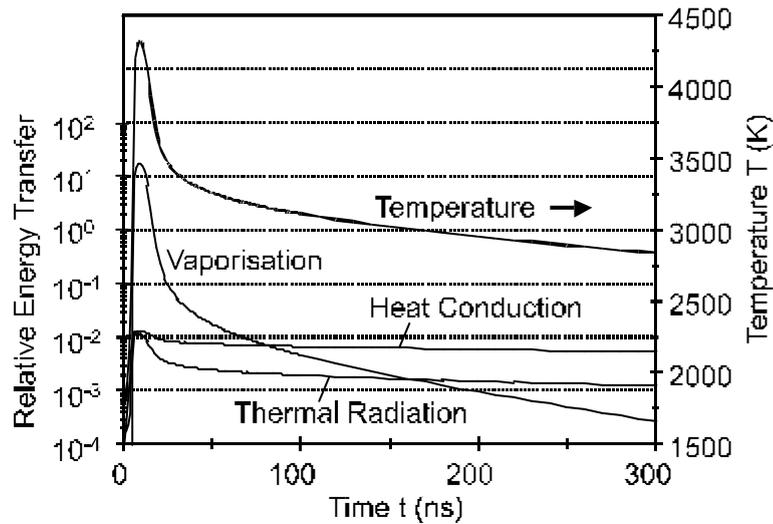


Fig 2.: Relative amplitude of the different heat loss mechanisms for a 30 nm particle.

With this approximation the maximum particle temperature and derived from this the maximum signal can be calculated analytically. According to Melton [1984], the signal for a given emission wavelength  $\lambda_{em}$  is thus given by

$$S_{LII} \propto N_p \cdot d_p^x, \quad (5)$$

where  $N_p$  denotes the local number density of primary particles and the exponent  $x$  is given by

$$x = 3 + \frac{154nm}{\lambda_{em}}. \quad (6)$$

As a result, the LII signal at the moment of maximum temperature can be used for the approximate measurement of the soot mass concentration if appropriate detection wavelength are chosen. It should be pointed out that equation 6 yields a slight overestimation of larger particles, which might be tolerated for most applications. The proportional constant in equation 5 given by the individual experimental set-up may be obtained by an simple line-of-sight extinction measurement [Will et. al., 1996].

### Experimental aspects for in-cylinder soot measurements

The application of LII for in-situ measurements of soot mass concentration within the combustion chamber of a diesel engine, however, requires some additional aspects to be considered in order to obtain reasonable results.

#### Excitation and detection wavelengths

One important point is to reject background luminosity of soot particles outside the probe volume. Although the thermal radiation of laser heated particles is orders of magnitude higher than the radiation of flame soot at typical combustion temperatures of about 2000 K, the line-of-sight integration of this background signal may possibly be a major source of interference in LII experiments. However, by a careful choice of the detection wavelengths range a sufficient suppression of this effect can be realized. This can be clearly seen from figure 3, where the quotient of the signal ratio of LII and the ambient flame radiation is displays as a function of detection wavelength. As a result, at short detection wavelengths a better background rejection is feasible [Dec, 1992]. This is due to Wien's displacement law which yields a blueshift of the wavelength of maximum emission for increasing temperature. At a detection wavelength of 450 nm, marked within the figure, a suppression ratio of about 3000 is achieved.

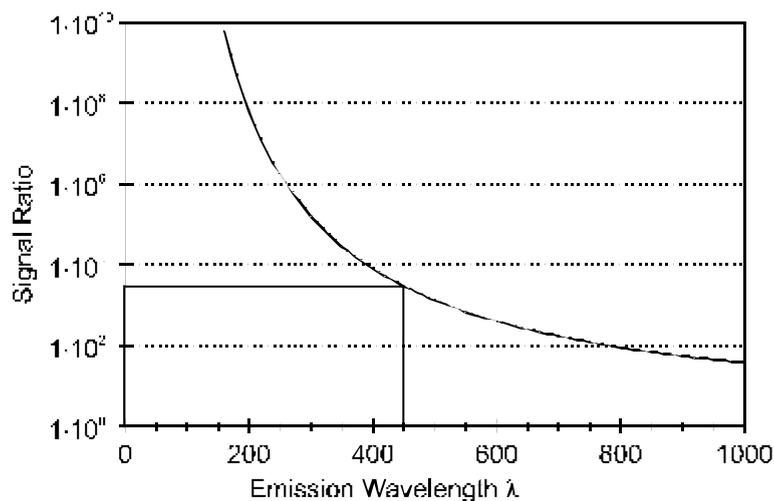


Fig 3.: Ratio of blackbody spectral radiant exitance of laser heated soot (4000K) and flame soot (2000K)

Another aspect which has to be taken into account is the avoidance of molecular band luminescence, especially of chemically excited  $C_2$  molecules vaporizing from the particle surface. This effect is a particularly important source of noise, especially for high values of irradiance. The spectral range of these strong transitions

starts from about 470 nm [Pearse et. al., 1976]. Hence, this aspect suggests detection wavelengths blueshifted relative to 470 nm.

Also the interference of elastically scattered light from particles and surfaces has to be suppressed. This is, of course, particularly important for the application of LII within the combustion chamber of optically accessible engines. For this reason, also a blueshifted detection range as compared to the incident laser excitation has to be chosen, or the other way round, a laser wavelength larger than about 500 nm should be selected. This selection has the additional advantage that there is no interference of fluorescences which might be excited by the laser pulse. However, no excitation wavelength in the infrared should be used, as the absorption efficiency is strongly wavelength dependent and much higher radiant fluxes are necessary here. Therefore, a good selection is given by the application of a frequency doubled Nd:YAG laser at a laser wavelength of 532 nm, which is easy to handle and provides suitable pulse energy within reasonable pulse durations (about 8-10 ns).

However, it should be considered that for high signal values long detection wavelength have to be preferred, according to equation 3. Additionally, the mass proportionality of the LII signal generally is only given for detection wavelengths as long as possible, which can be seen from the equations 5 and 6.

A good compromise of all aspects mentioned above, however, depends on the individual experimental conditions. From equation 6 the goodness of the mass proportionality can be estimated by calculating the exponent  $x$  for the given detection wavelength range.

#### *Extinction of laser beam and signal*

One general problem for optical measurement techniques within technical combustion system arises from the extinction of light. Besides absorption and scattering of light by particles and droplets within the optical path, blinding of windows and mirrors are the major source of laser and signal attenuation. A discussion of these effects for LII measurements, however, has to be done separately for the incident beam and the signal.

In contrast to linear measurement techniques, like Mie scattering, the power dependence of the LII signal shows a saturation behavior which can be used for a minimization of laser beam attenuation effects. In figure 4 the power dependence of the LII signal is given for a 30 nm particle. From this graph, it can be clearly seen that after an initial linear range there is a saturation of the LII signal for values of the laser irradiance starting at about  $10^8$  W/cm<sup>2</sup>. This is due to the onset of vaporization which limits a further temperature increase and even results in a signal decrease because of particle shrinking. However, this behavior can be utilized for a reduction of beam attenuation effects. If an appropriate value of laser irradiance is chosen there is almost no effect on the signal along the laser path, even if strong attenuation of about 50% is present. Even less severe is the effect of window blinding as the pulsed laser itself is continuously cleaning the windows by vaporizing condensed humidity and particles.

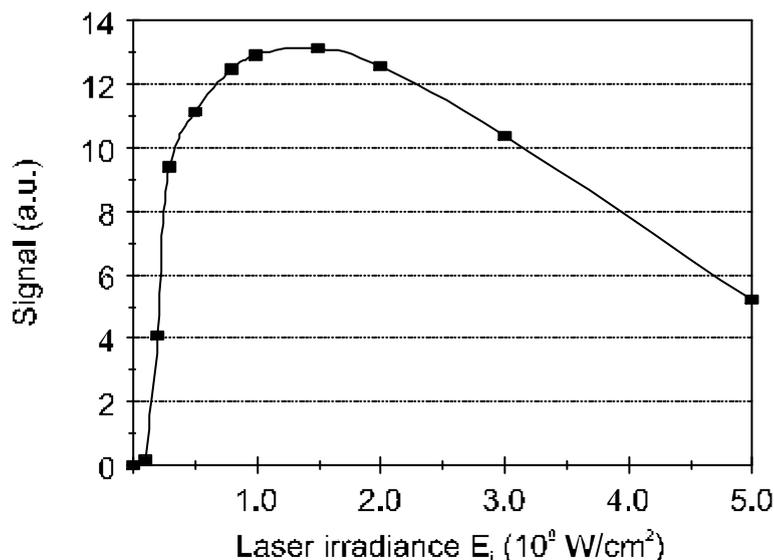


Fig 4.: LII signal as a function of laser irradiance for a 30 nm particle.

However, signal attenuation is much more difficult to prevent. According to Lambert-Beer's law there is a signal decrease depending on the droplet and particle concentration within the signal path due to absorption and scattering. Therefore, the view direction should generally be chosen to prevent passing dense regions like the injected spray or highly sooting clouds. Additionally, soot condensation at the detection windows has to be minimized. For the synchronization of the engine and the laser repetition rate, it has to be justified that fuel injection and ignition is only present within these engine cycles where actually laser irradiation occurs. This includes the acceleration of the engine to the desired speed with an electrical generator, which has, however, the disadvantage, that no thermal stationarity of the engine is achieved. A continuous cleaning will always be necessary, regardless of additional measures to minimize window blinding. Depending on the specific operating conditions this should be done at least after about 20 injections.

### Experimental set-up

For this study, the object of investigation was an optically accessible DI diesel engine on basis of a MAN D0824 LFL 06 series engine. In order to ensure realistic conditions, the standard crank case and cylinder head was applied with almost no modification. In-between, an elongation piece was applied to take of the elongated piston which is mandatory for optical access through the piston bowl. By this construction (figure 5) a compression ratio of  $\varepsilon=16.7$  (serial standard  $\varepsilon=18.0$ ) is achieved and the intake air flow and injection system remains unchanged. However, the geometry of the piston bowl [Münch et. al., 1997] has to be modified for optical access. An summary of the engine specifications is given in table 1.

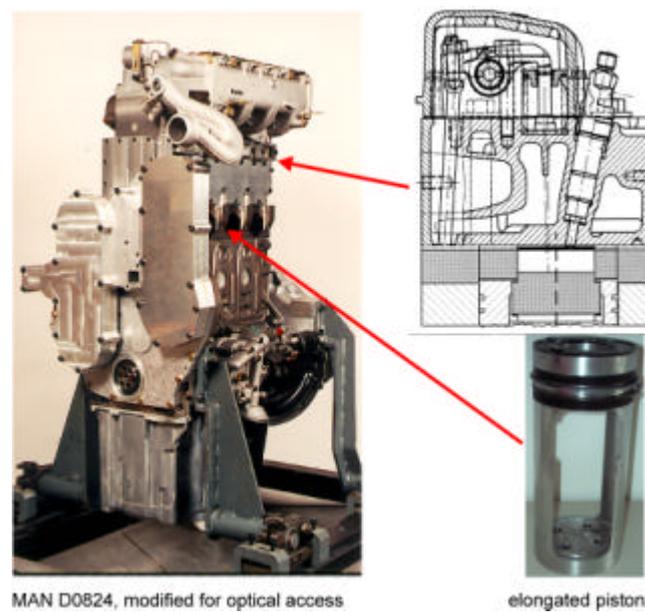


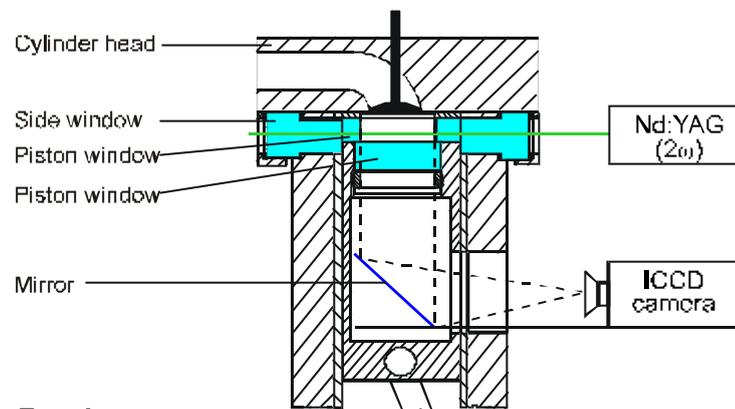
Fig 5.: Optically accessible engine on basis of MAN D0824.

For two-dimensional measurements, side windows were applied for the incident laser sheet, which was horizontally aligned and coupled into the piston bowl through a glass ring, applied as a transparent piston crown. The LII signal was detected through a transparent flat bottom piston bowl. The light sheet was imaged on an intensified high speed charge-coupled-device camera (ICCD) over a mirror, which was placed into a slit within the piston (figure 6).

Table 1: Engine specifications

Engine Manufacturer	MAN Nutzfahrzeuge AG	Compression ratio	$\epsilon=16.7$
Engine Type	D0824 LFL 06, 4-cylinder in-line	Engine speed	1000 rpm
Combustion system	common-rail diesel direct injection, turbocharged	Injection pressure	60 MPa
Displacement/cylinder	1145 cm <sup>3</sup>	Nozzle type	7 hole Sac-hole nozzle, (Bosch DLLZ)
Bore $\times$ Stroke	108 mm $\times$ 125 mm	Start of injection	5° CA before TDC
Diameter of piston bowl	60.5 mm	Injected fuel volume	60 mm <sup>3</sup>

**Side view**



**Top view**

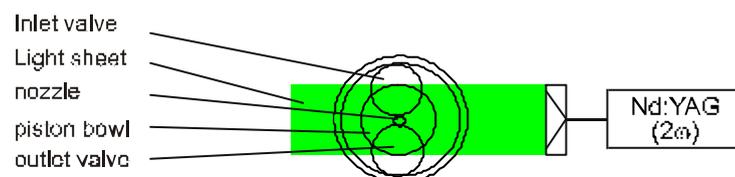


Fig 6.: Schematic of experimental set-up.

The engine was driven on a standard engine test bench by an electrical generator ( $P=50$  kW). Hereby, only one out of the 4 cylinders was fired. The injection parameters have been fully controllable and injection was only started after stationarity of the desired engine speed. By an increased boost pressure (247 kPa) and intake air temperature (335K) the serial values of the compression pressure (6.5 MPa) and air mass were realized, despite of the decreased compression ratio of the modified engine. This can be seen from figure 7, where the cylinder pressure as a function of crank angle is displayed for the investigated operation conditions.

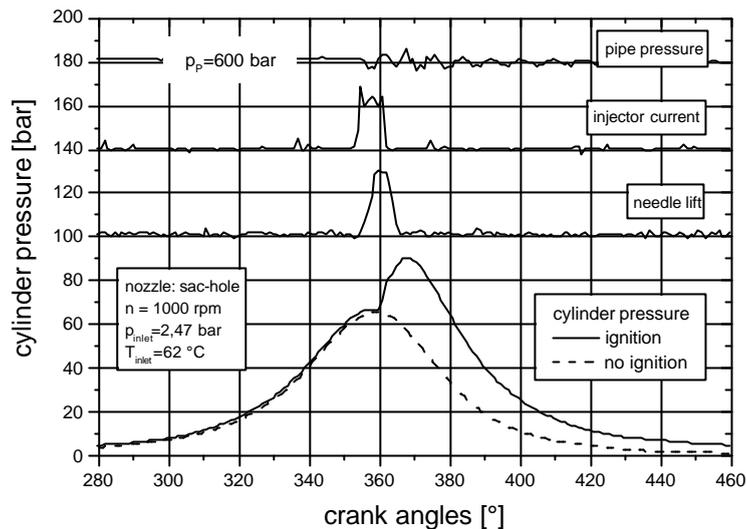


Fig 7.: Indicator diagram of cylinder pressure, pipe pressure, injector current and needle lift.

By an appropriate trigger unit the laser was synchronized to the engine keeping the repetition rate in an interval of about 9.5 to 10.5 for stable operation conditions. For this purpose, some engine cycles have to be omitted for observation. To minimize window blinding, no fuel was injected within these cycles.

## Results and discussion

As a major problem, soot condensation on the piston bowl window was observed, which also makes a calibration for quantitative measurements, which was not performed within this study, more complicated. It has been observed that, with an increased number of fired cycles, there is a soot layer on the bottom of the piston bowl, which is continuously growing. After about 20 single injections, the piston bowl window has to be cleaned to ensure the comparability of different individual images.

The detection time was varied in steps of 1° CA around top dead center and in steps of 2° CA later on to obtain the temporal evolution of soot formation and oxidation within the combustion chamber. As the laser repetition rate is limited to about 10 Hz, the individual images within these sequences were observed in different engine cycles and therefore no real evolution of flame structures can be investigated, but the amount and location of occurrence of soot within the detection plane can be compared for different detection times. In figure 8 a sequence of the soot mass concentration for 1000 rpm and 60 mm<sup>3</sup> injected fuel is depicted.

It can be observed that locally areas of high soot concentration are present within the detection plane. However, the location and concentration are statistically distributed due to cyclic fluctuations. Additional information about the internal combustion process is accessible by a simultaneous measurement of the flame luminosity, which is detected within the same spectral range as the LII signal, but without laser irradiation and with much larger detector gates than for LII experiments. From these measurements also a sufficient suppression of background luminosity can be verified.

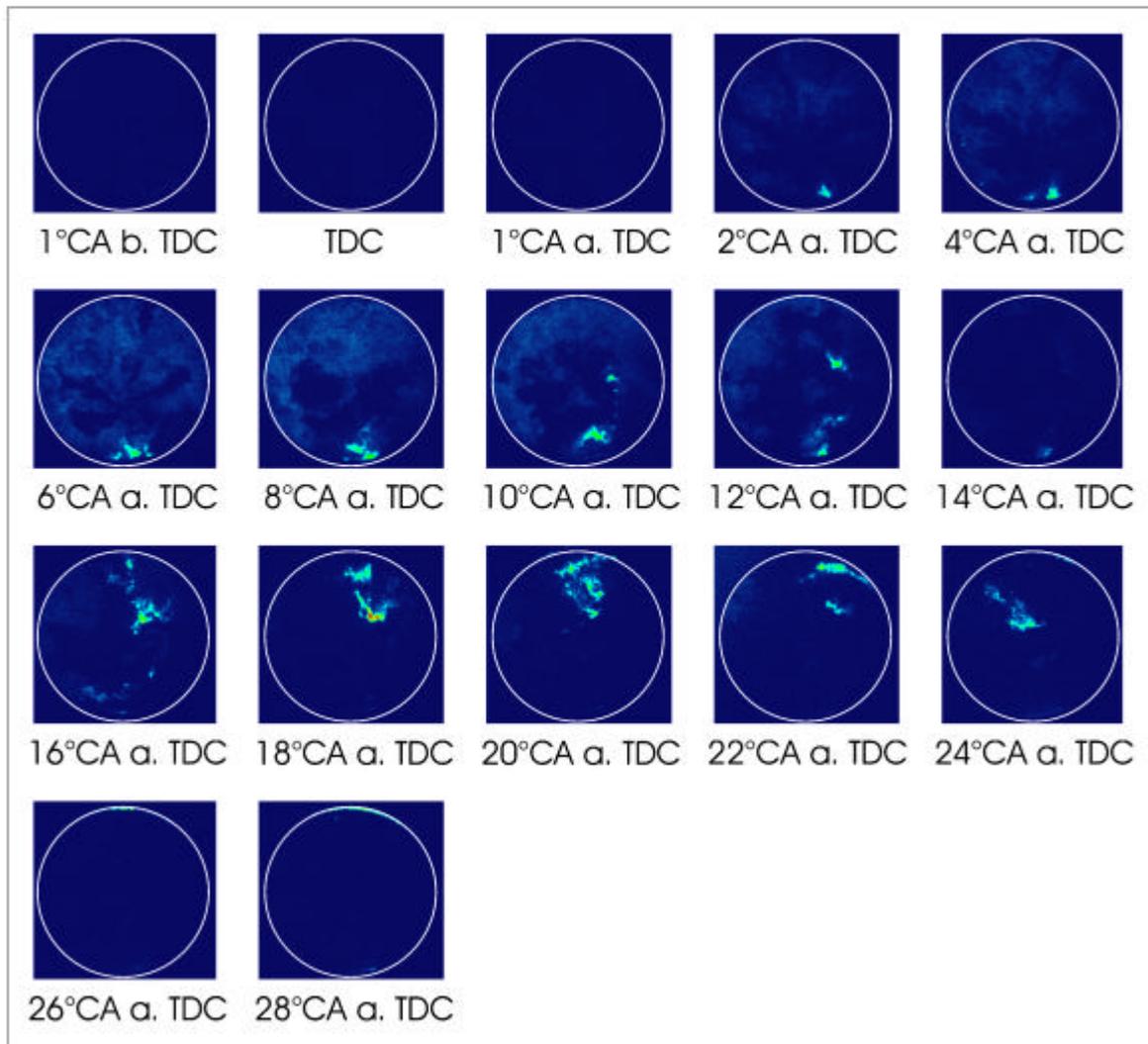


Fig 8.: Temporal sequence of soot mass concentration within the piston bowl of an optically accessible engine.

From a comparison of the LII signal to flame luminosity measurements (figure 9) it can be seen that the first occurrence of the LII signal for a detection time of 2° CA after TDC temporally coincides with the start of the diffusion flame. After the end of the diffusion flame, which is given at about 18° CA after TDC, there is still a significant LII signal observable. The corresponding cold soot will not be further oxidized and will be emitted during the exhaust stroke of the engine. In contrast to LII measurements much less fluctuations of the flame luminosity have been observed. The main reason is given by the fact that the luminosity signal is integrated over the whole depth of the piston bowl, whereas the LII signal arises only from a thin light sheet with a thickness of about 500  $\mu\text{m}$ . This integration blurs finer structures of the flame. The LII signal is, however, only present, where the flame front crosses the detection plane. Therefore, a detailed investigation of the locations of soot formation is possible by scanning the laser beam across the complete depth of the piston bowl. This information can not be gathered by the detection of flame luminosity. Additionally, cold soot which is present after the end of the diffusion flame is only detectable by LII measurements.

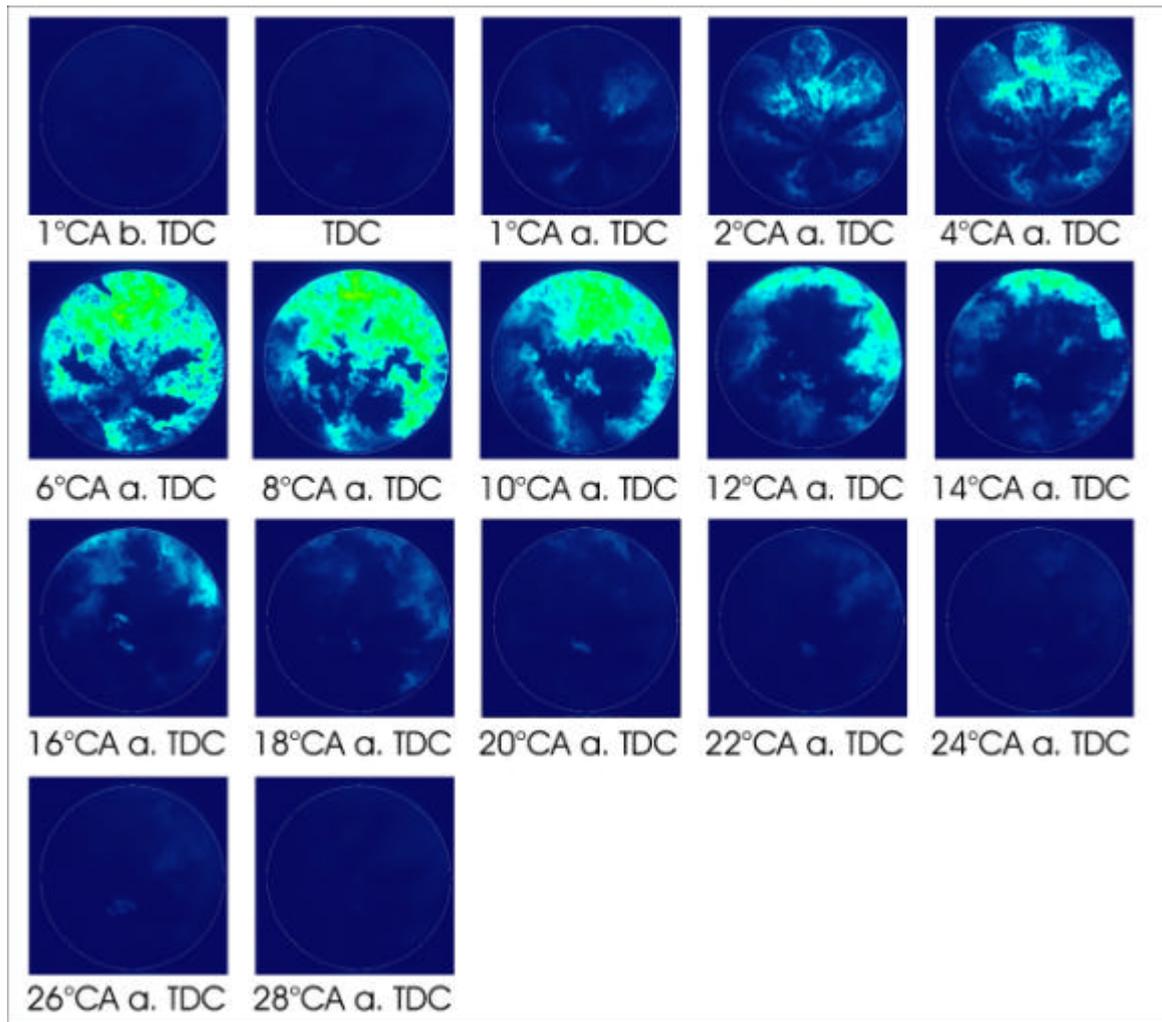


Fig 9.: Temporal sequence of flame luminosity within the piston bowl of an optically accessible engine.

## Conclusions

The LII measurement technique has been successfully applied to a series engine, modified for optical access. The soot mass concentration within a thin light sheet has been determined without interference. By an appropriate selection of the excitation and detection wavelengths, a complete suppression of elastically scattered light from walls, dust and droplets as well as a good rejection of background luminosity is achieved. It has been shown that by a simultaneous detection of flame luminosity, additional information can be obtained.

First results are shown for a heavy duty truck engine, where necessary modifications have been performed in a manner to meet the series engine specifications as close as possible. The decreased compression ratio is compensated by an increased boost pressure at an increased intake air temperature. With this measure, almost realistic engine conditions are given except for the piston bowl geometry. In this study, soot formation of standard diesel, injected by a sac-hole nozzle was investigated and discussed for different observation times within the engine cycle. It has been observed that also high soot concentrations are found for late detection times after the end of the diffusion flame. This portion will likely not be further oxidized and later on emitted within the exhaust stroke.

Future applications will have to focus on the minimization or prevention of window blinding and on the quantification of the soot mass concentration obtained by this technique.

## Acknowledgements

The authors gratefully acknowledge financial support of parts of the work by the Deutsche Forschungsgemeinschaft (DFG), the Bayerische Forschungsförderung (BFS) and the companies AUDI AG, BMW AG and MAN Nutzfahrzeuge AG in the frame of this cooperation.

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