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On-line Diesel soot diagnostics by time-resolved laser-induced incandescence (TIRE-LII)

On-Line Diesel soot diagnostics by time-resolved laser-induced incandescence (TIRE-LII)

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Laser-induced incandescence (LII) is presented as a favorable optical measurement technique for a comprehensive Diesel soot characterisation in the combustion chamber as well as within the exhaust gas. Currently, for soot mass concentration measurements filter smoke meters, the opacimeters and some variants of them are used for most purposes, including inspection and maintenance. However, there are some deficiencies connected to these methods. In particular, for transient behavior, soot concentration is becoming more difficult to investigate, as samples have to be collected over a sufficiently long period of time to ensure accurate data. In contrast, optical techniques generally offer the possibility for real-time measurements, but extinction measurements and light scattering fail to produce accurate measurements for low soot concentrations.

Particle size measurements up to now focus primarily on the aggregate size as the investigated parameter. Usually, the mobility of electrical charged particles is analyzed with, e.g., differential mobility analyzers (DMA) or scanning mobility particle sizers (SMPS) [1]. If primary particle size has to be determined, sampling with subsequent transmission electron microscopy is currently in use as an ex situ technique [2]. However, a problem with these techniques arises from the fact that diesel exhaust gases additionally contain droplets of sulfuric acid, which may falsify the results and lead to misinterpretations [3].

Concerning these problems LII is a promising measurement technique, which has been used for soot diagnostics for the last ten years. Quantitative on-line measurements of both the primary particle sizes, based on the analysis of particle cooling, and the mass concentration are simultaneously feasible, even under very low sooting conditions [4-6].

The basic principle of laser-induced incandescence is to heat up the soot particles by a highly energetic laser pulse from ambient temperature and to analyze the enhanced thermal radiation. For high values of laser irradiance (some tens of MW/cm^2) the particles reach temperatures of about 4000 K within a few nanoseconds. For suitable detection wavelengths, the radiation increases by several orders of magnitude, and a clear separation of the signal from background luminosity is possible. The thermal radiation of the particles can be detected by an appropriate detector and subsequently be evaluated for mass concentration and particle size. For this purpose particle heating and cooling processes are modeled by setting up the power balance equation. The different terms of this equation, which is shown on slide 4 denote absorption of laser irradiation, heat loss due to conduction, vaporization and radiation and the change of internal energy, respectively. These mechanisms are discussed in more detail by Hofeldt [7], Melton [8] and Dasch [9].

In setting up this equation we assumed spherical primary particles which have only point contact and which can form aggregates of various size. For this morphology which is confirmed by transmission electron microscopy (TEM) for several objects of investigation and for a sufficiently loose structure, mutual interferences between the individual particles within a cluster can be neglected and the heat transfer is mainly determined by the specific surface of the particles and thus by the primary particle size d_p. The absorption cross section can easily be calculated by an algorithm including the Mie formulae [10], because an ensemble of spherical particles can well be treated as isolated spherical particles in this case [11, 12].

By a numerical solution of the power balance, the temporal profiles of particle temperature and size can be derived and model curves for the signal decay can be obtained by integrating the spectral radiant excitance weighted by the spectral response of the detector, given by

$$S_{LII} \propto d_p^{\,2} \int R(\lambda)\varepsilon(\lambda, d_p) M_\lambda^{\,b}(T,\lambda) d\lambda \text{ (slide5).}$$

It can be shown that for high laser irradiance at the moment of maximum temperature the signal is nearly proportional to the local soot volume fraction if suitable detection wavelengths are chosen.

If quantitative values for the mass concentration are required it is necessary to perform an appropriate calibration. This can easily be done by a single line-of-sight extinction measurement [4]. Therefore the light of a continuous-wave laser is passing through the detection volume and the total transmission yields the soot volume fraction integrated along the path. From this a calibration factor can be derived which allows quantitative measurements of the mass concentration for all operating conditions.

Additionally to the applicability of LII to concentration measurements, the temporal behavior of the signal can be analyzed. Conductive cooling is the dominant heat loss mechanism for late detection times and it is, at constant ambient temperature, only a function of specific surface and thus of primary particle size. In previous work [13-14] we showed that the primary particle size can be obtained by calculating model signal decays for the given experimental conditions and comparison to experimental data. As only relative signal changes are evaluated this approach directly yields the desired parameter without the need for further calibration. As a specific example in slide 6 the normalized signal decay is depicted for various primary particle sizes, which are typical for raw diesel exhaust gases.

LII measurements were also performed inside the combustion chamber of an optically accessible engine using the experimental set up shown on slide 7. The piston has glass windows for transmission of a laser light sheet and also a glass bottom for detecting the LII-signal, which is reflected by a mirror onto an ICCD-camera. Thus it is possible to obtain two-dimensional information in a imaging way from only one single laser pulse. Exemplarily, a temporal sequence of soot concentration is shown for a Common Rail truck engine (slide 8).

The pointwise measurements in the exhaust gas of a passenger car Diesel engine, cf. also [6], were performed about 60 cm behind the manifold and the turbocharger within the exhaust gas where it spread out into ambient air without any further exhaust aftertreatment and without any exhaust silencer. The beam of a frequency doubled Nd:YAG-laser operating at a wavelength of 532nm was used to illuminate the exhaust gas perpendicular to the gas flow directly at the end of the pipe without any further focusing optics. The LII signal was detected perpendicularly to the direction of the incident beam and to the gas flow with a fast photomultiplier tube module (PMT), again without any focusing lens (slide 9). The detector was placed at a distance of about 25 cm from the sample volume, and an appropriate wavelength range was chosen by a filter with exactly known spectral characteristics.

Under spatially homogenous conditions, as given within the exhaust pipe, pointwise information about primary particle size can be obtained by the evaluation of the full signal decay. The temporal response of the detector was monitored with a fast digitizing oscilloscope and evaluated by a personal computer.

The soot mass concentration was calibrated with a single line-of-sight transmission measurement, which has been performed with a HeNe-laser at a wavelength of 632.8 nm. Extinction was measured by a photodiode, which in combination with the adjusted absorption length directly yields the mass concentration. By this, a calibration factor was determined at highly sooting operating conditions, which allows quantitative measurements even at very low sooting conditions, which are generally not accessible with extinction measurements.

As it is an important input parameter for Time-Resolved(TIRE)-LII measurements, the temperature of the gas flow was continuously measured with a thermocouple.

At each operating point the temporally resolved signal curve was averaged over 20 single laser pulses in order to reduce statistical noise and, even more important, temporal variations within individual cycles. As a specific example the corresponding curve for 1000 rpm and 6 bar p_{me} is depicted in slide 10.

The soot mass concentration within the raw exhaust of the engine is shown in slide 11. It can clearly be seen that there is a significant decrease of the soot mass concentration with increasing engine speeds and, for the two higher engine speeds, towards larger mean effective pressure.

This might be due to the fact that the mixture formation is more homogeneous within the engine at higher engine speeds, inducing higher turbulence rates of the gas flow. With increasing mean effective pressure the combustion temperature is increasing due to the higher amount of injected fuel. As a consequence, the NO_x concentration is increasing, which was also measured, and the soot mass concentration decreasing because of more effective oxidation.

As expected, there is a dramatic increase of soot emission observable for cold start conditions. The reason for this might be either that particles are larger or that there are more particles produced.

The answer can be found from the simultaneously measured primary particle sizes, which are shown on the left side of slide 12. The values are obtained from the same signal curves as for the mass concentration by evaluating the decay time constant derived from a single exponential fit of the experimental decay. The standard deviation in this case was about 1 nm for all conditions. An additional data point is added for the size which was determined by TEM. Within experimental uncertainties the agreement found is excellent.

The primary particle size is nearly unaffected by the fact whether cold start conditions are chosen or not. So the higher soot mass concentrations are caused by an increase of the number of primary particles whereas the properties of the single particles seem to be unaffected. This behavior can clearly be seen in the right figure on slide 12, where the number concentration of soot primary particles is depicted. The conclusion drawn from this result is that under cold start conditions the number of condensation nuclei is increasing, whereas surface growth seems to be affected only marginally. Furthermore, it can be seen that the primary particle size in general decreases with increasing engine speed. This behavior might be due to the fact that reactions for surface growth have less time to occur and possibly lower PAH concentrations additionally slow these processes. For a variation of the mean effective pressure no general trend can be found.

It has been demonstrated that laser-induced incandescence is an appropriate tool for accurate soot emission measurements both inside the engine and in the exhaust gas. Simultaneously accessible quantities are the mass concentration, the primary particle size and, as a quantity derived from these, the number concentration of primary particles.

The technique described requires only little experimental effort, but produces data with high reproducibility. One major advantage is particularly the high temporal resolution of pointwise measurements, which makes it, in contrast to other established methods, possible to investigate also the transient behavior of the engine. For example, also cycle resolution is feasible for each cylinder separately, if measurements are performed directly behind the outlet valve. Generally, the technique is applicable at every location within the exhaust system and, as a further example, enables also the in situ investigation of important characteristics of oxidation catalysts and soot filter systems.

For doing this, the high sensitivity is an important feature. The minimum mass concentration detectable by LII is estimated to be in the order of 100 $\mu g/m^3$ and even lower, if optics with large light collection are used.

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Outline

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- Motivation
- Laser-Induced Incandescence (LII)
- Investigations of soot inside an engine combustion chamber
- Soot characterisation in exhaust gases
- Conclusions

Motivation

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Benefits of laser-induced Incandescence for Diesel soot characterisation :

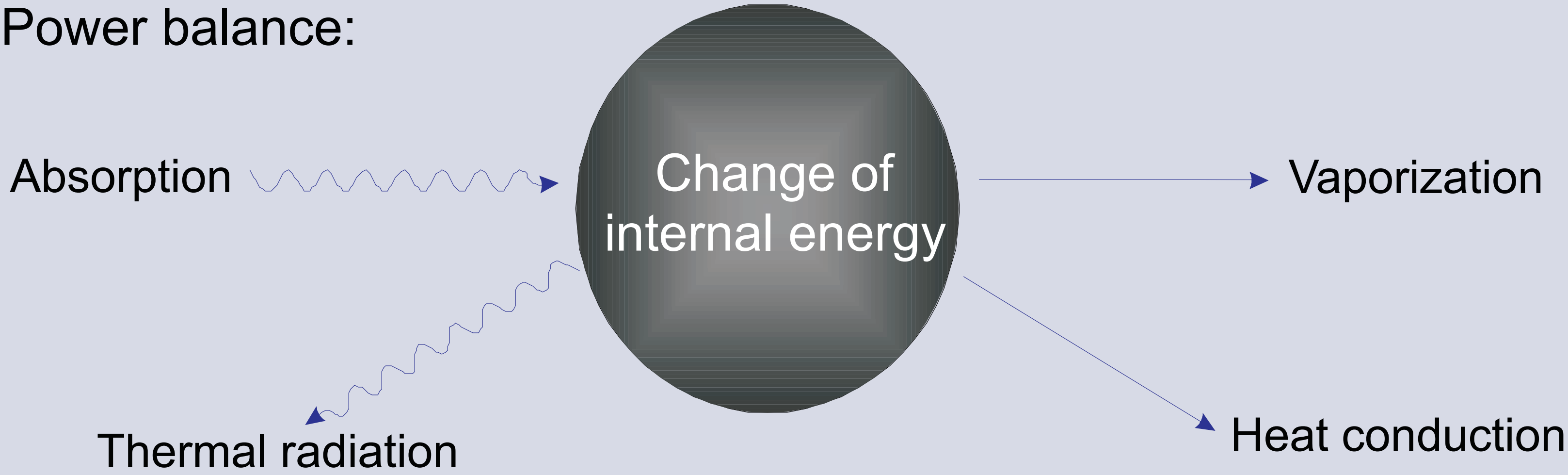
- Comprehensive characterisation of carboneous exhaust particles :
  - mass concentration
  - particle size

  } number concentration
- Measurement of carbonaceous particles, without interferences from condensates, sulfur, ...
- Increased sensitivity, particularly for ultra-low-emission engines (applicable for all possible test cycles)
- High temporal resolution, applicability for transient measurements (better 1Hz, acceleration, ETC ...)
- Non-invasive, no morphological change of the particles
- Easy adaptation, no need for a dilution tunnel

Principle of LII

Particle heating by means of a highly energetic laser pulse → Detection of enhanced thermal radiation

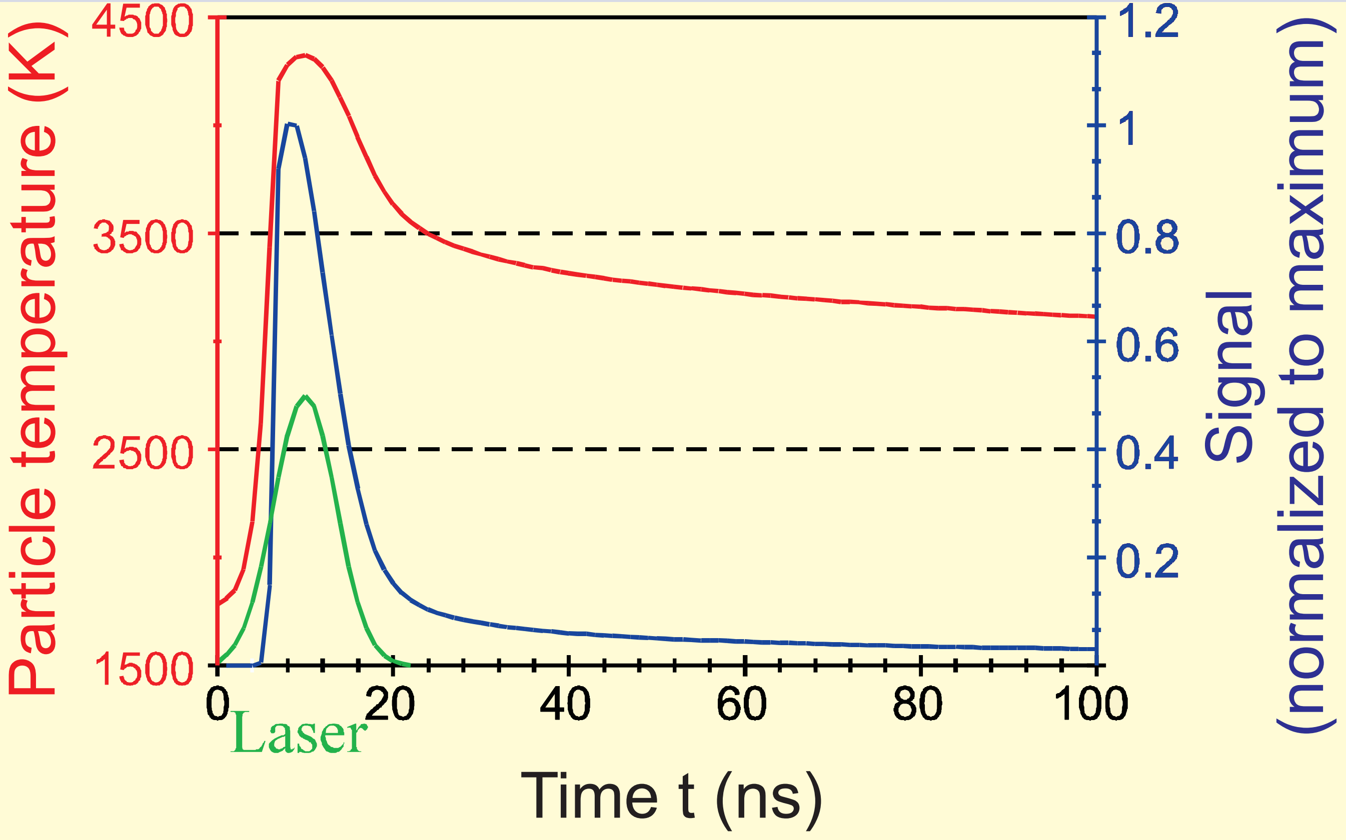
Power balance:



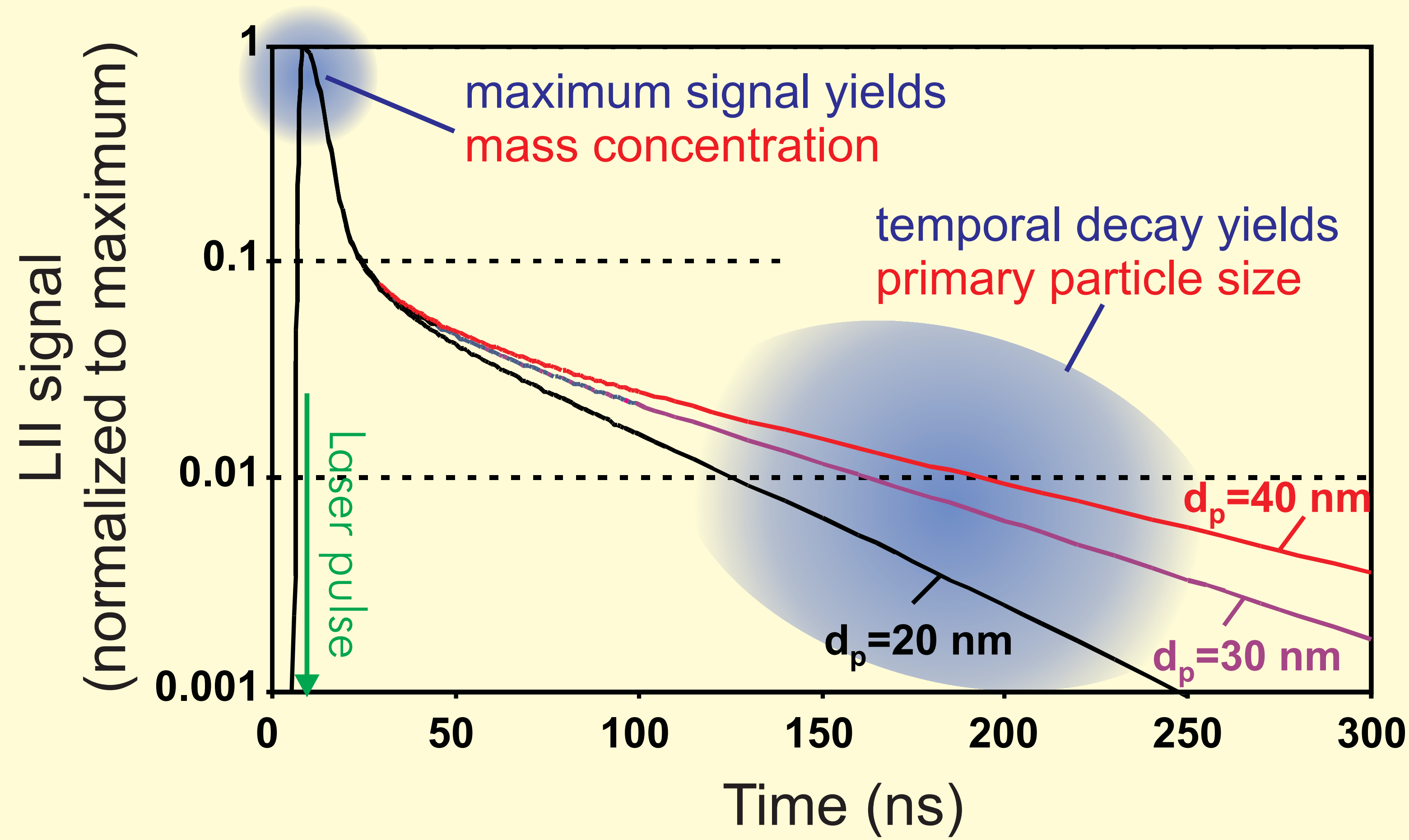
$$\underbrace{Q_{abs}\cdot\frac{\pi d_p^{\,2}}{4}\cdot E_i}_{\text{Absorption}} - \underbrace{\Lambda\cdot(T-T_0)\cdot\pi d_p^{\,2}}_{\text{Conduction}} + \underbrace{\frac{\Delta H_v}{M}\cdot\frac{dm}{dt}}_{\text{Vaporization}} - \underbrace{\pi d_p^{\,2}\int\varepsilon(d_p,\lambda)\,M_\lambda^{\,b}(T,\lambda)\,d\lambda}_{\text{Thermal radiation}} - \underbrace{\frac{\pi d_p^{\,3}}{6}\rho C\frac{dT}{dt}}_{\text{Change of internal energy}} = 0.$$

Principle of LII-Evaluation

Numerical solution of power balance yields temporal signal behaviour

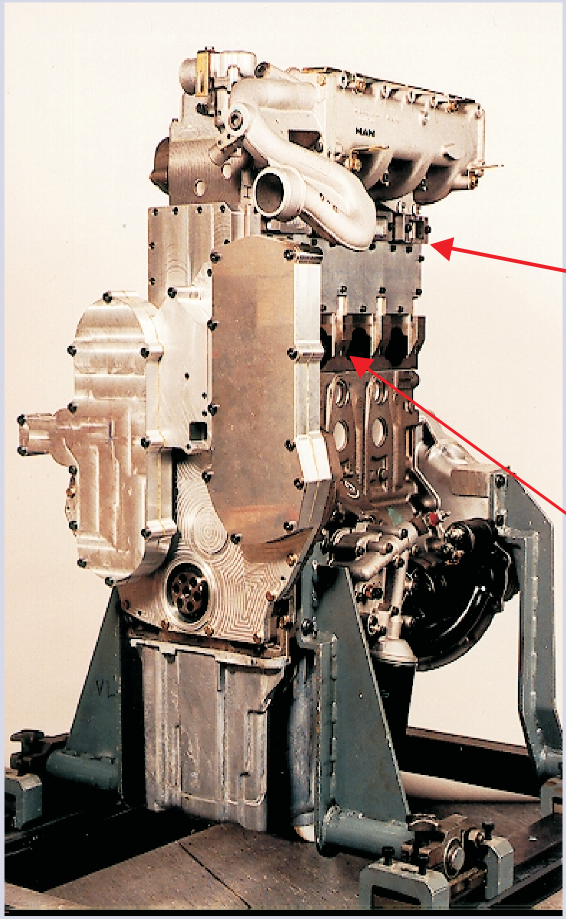


Quantities of interest

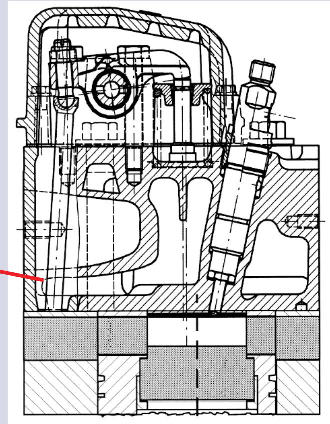


maximum signal in combination with elastic scattering ⇒ aggregate size

Experimental setup for LII in the combustion chamber

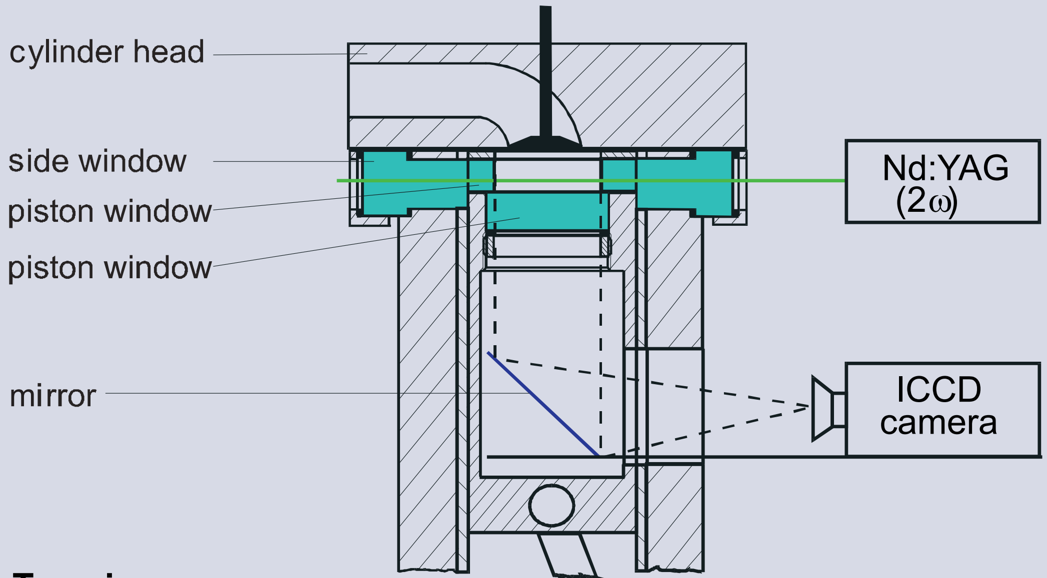


MAN D0824, modified for optical access

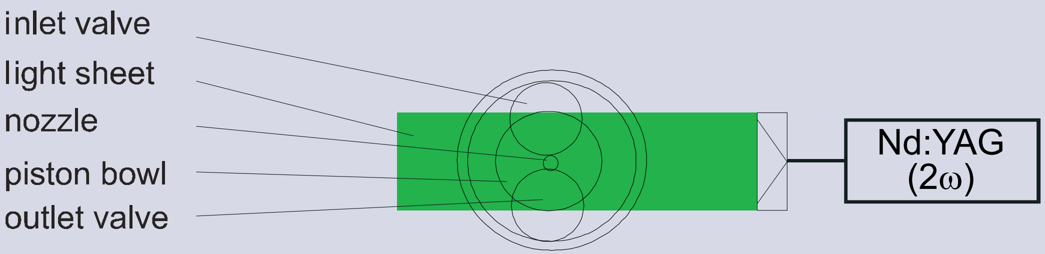


elongated piston

Side view



Top view



Temporal sequence of soot concentration

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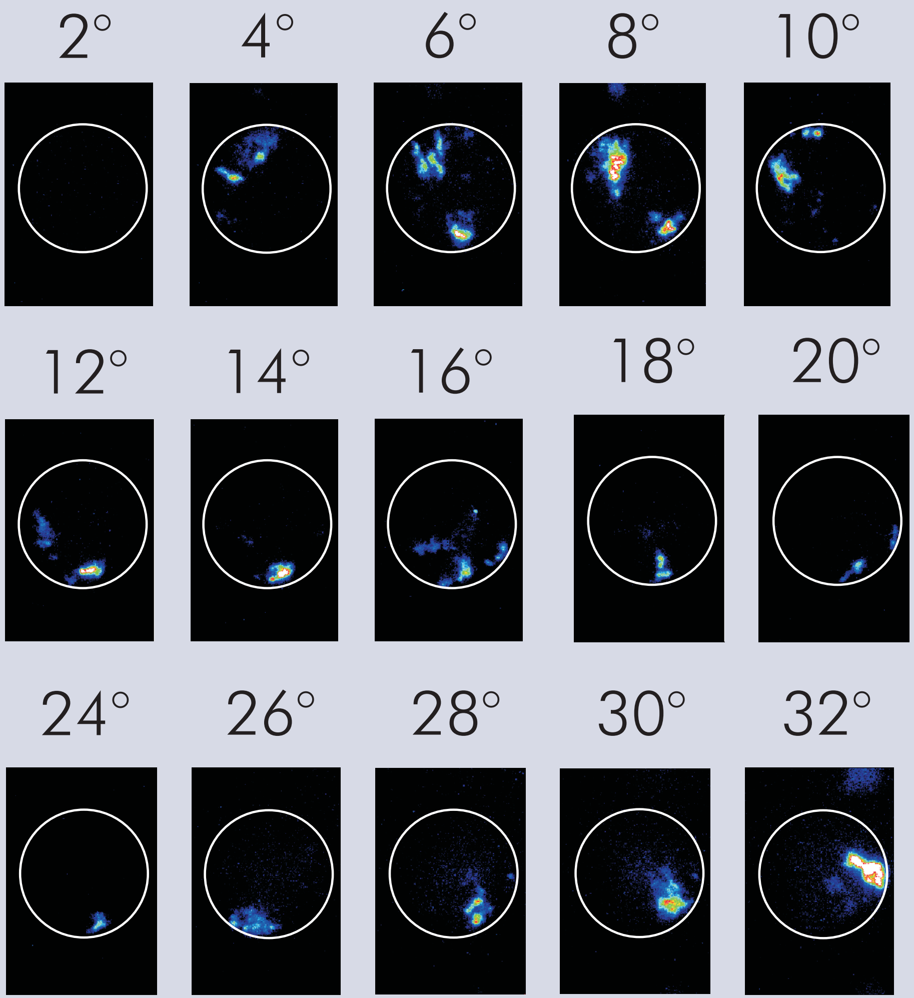


Time in CA after TDC

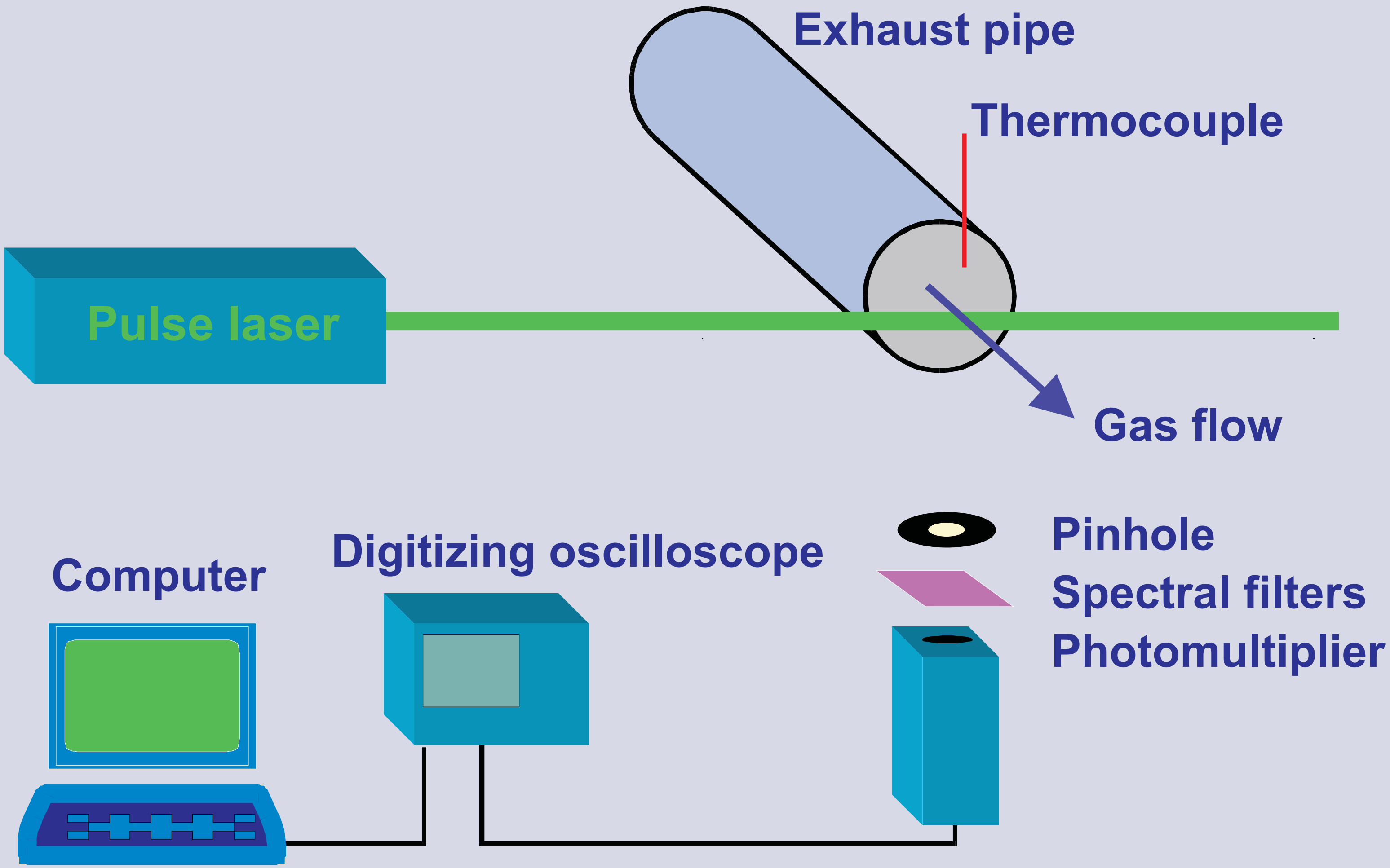
First LII signal at 4°CA after TDC

Strong cyclic fluctuations, small areas with particularly high soot concentration

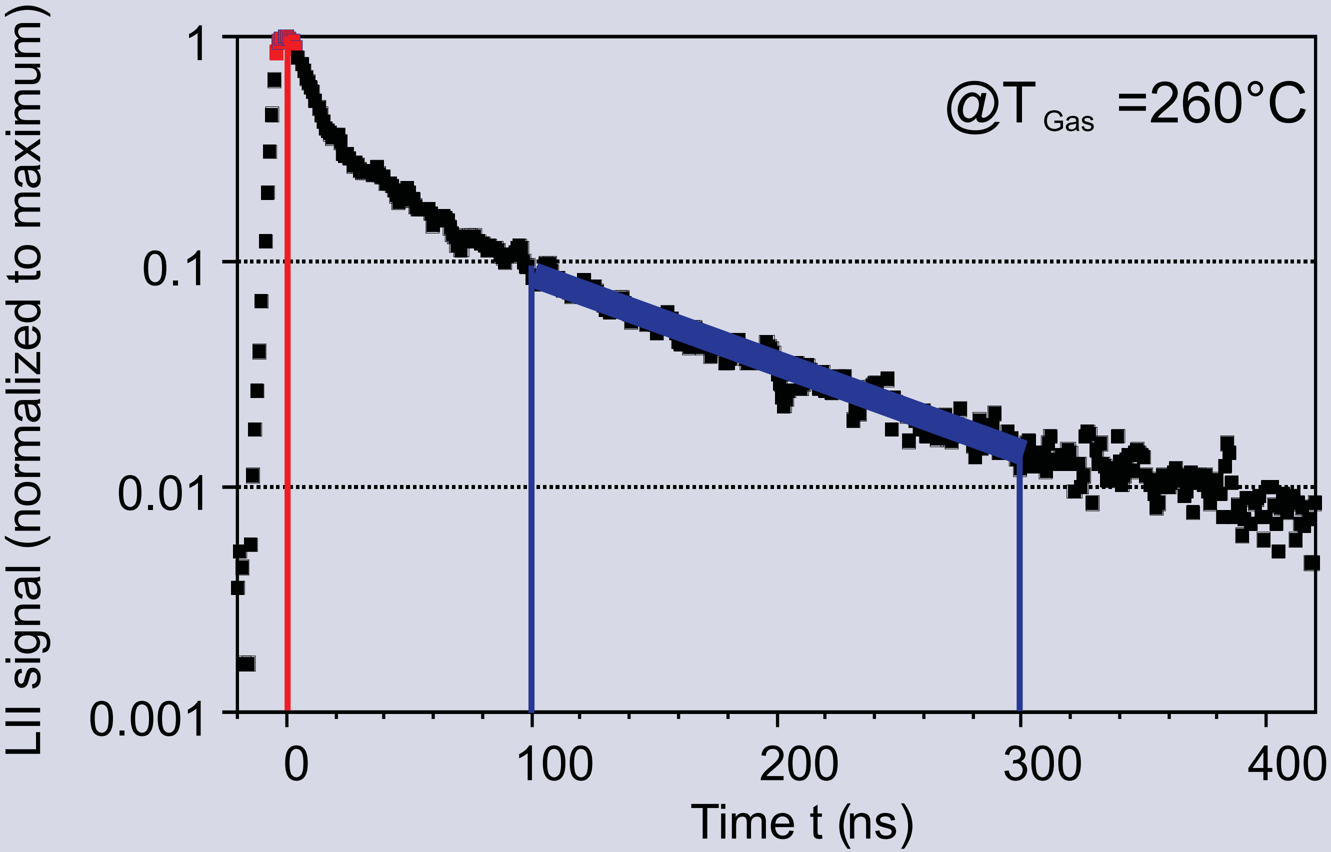
LII is even observable after the end of the visible flame

Two-dimensional signal, $d_{light\ sheet} \approx 500 \mu m$

Schematic set-up for exhaust gas measurements



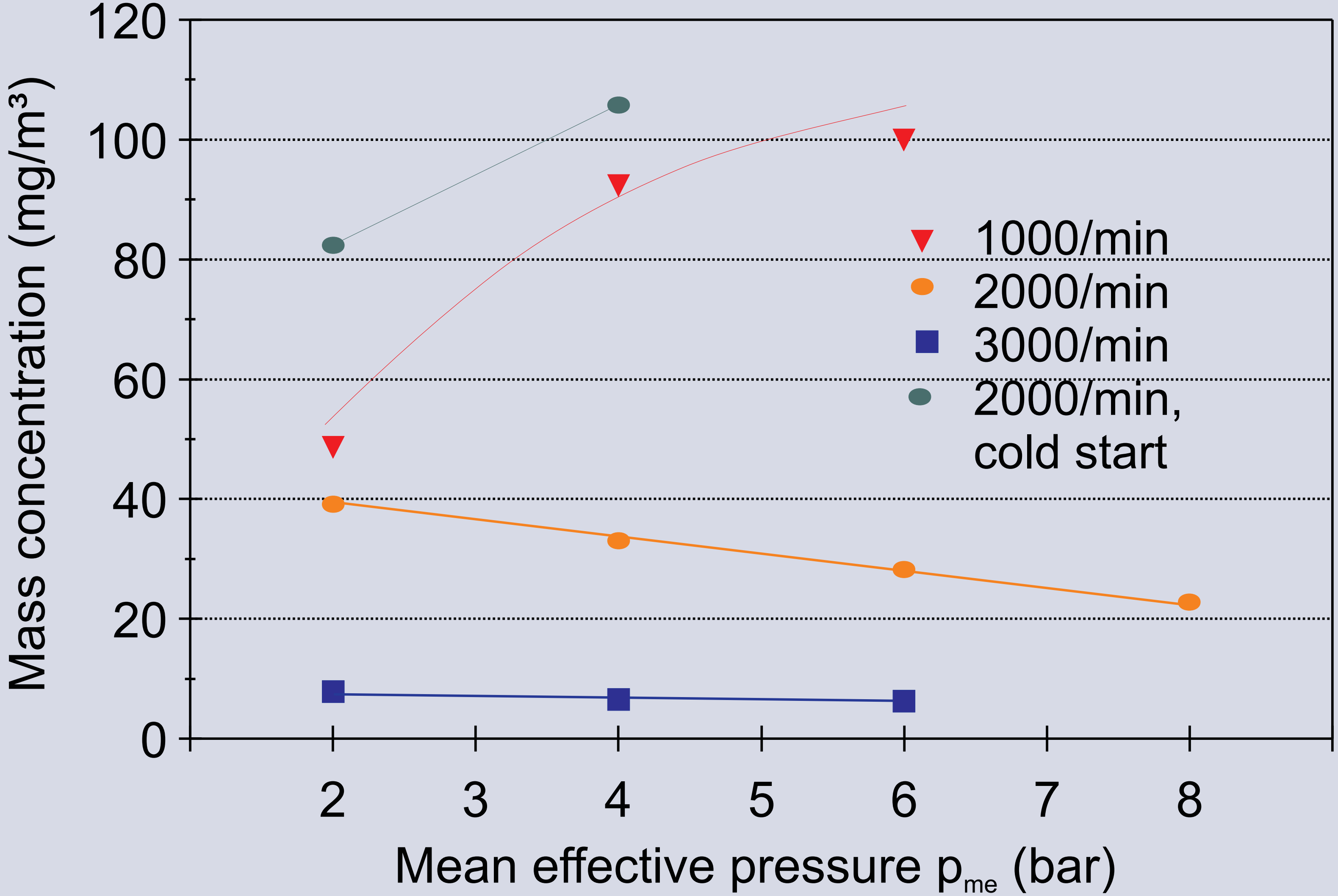
Example of experimental signal



- Decay time : ⇒ d_p=34 nm
  no calibration necessary, ambient temperature as an input parameter
- Signal heigth ⇒ m/V= 100,1 mg/m^3
  unique calibration necessary, e.g., by single line-of-sight extinction measurement

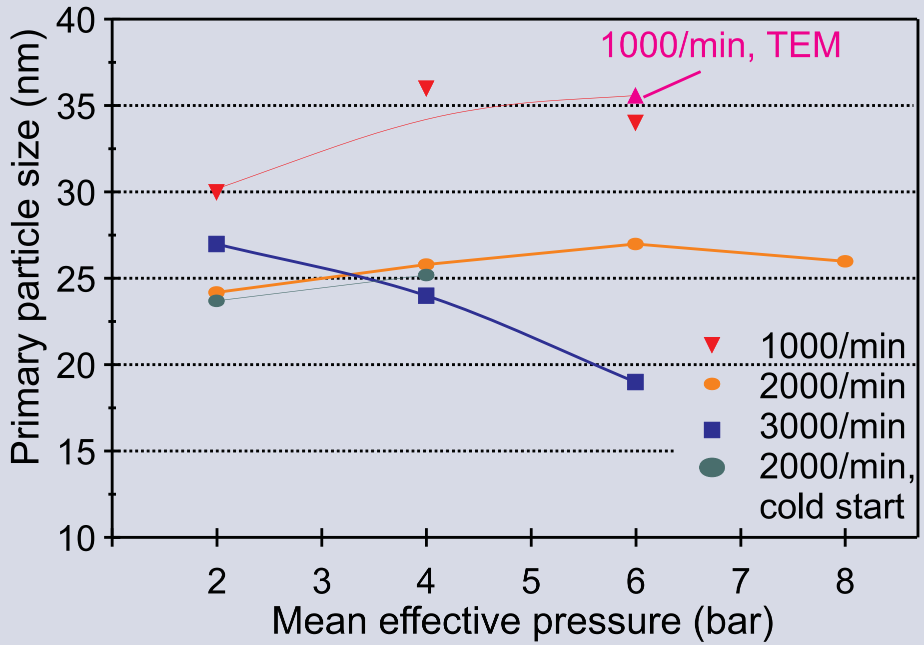
Results

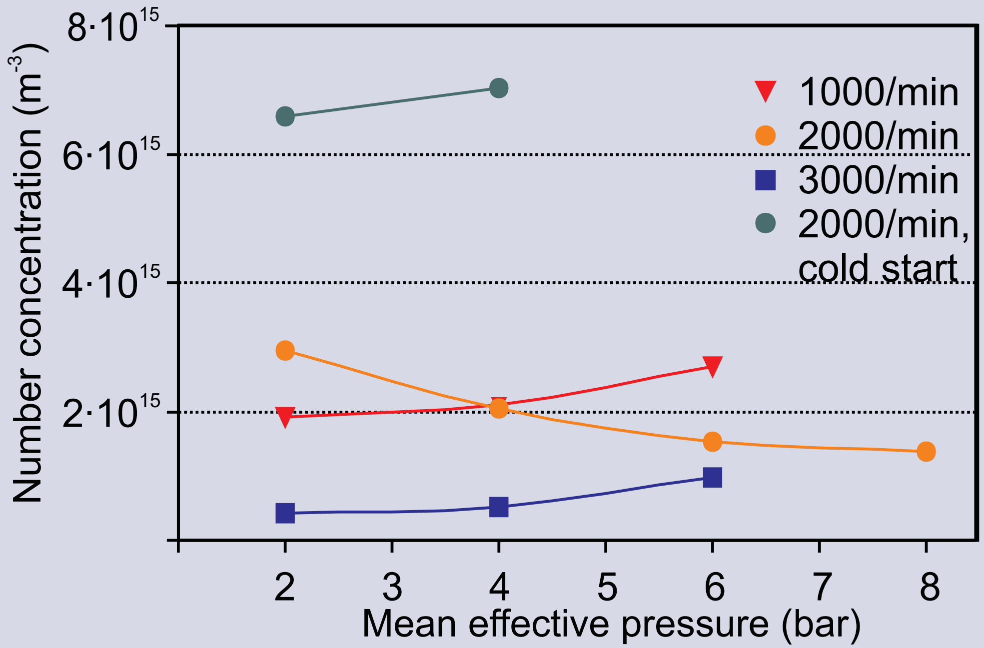
- Soot mass concentration (1,9l TDI Diesel passenger car engine)



Results

- Primary particle size and number concentration (1,9l TDI Diesel passenger car engine)





Conclusions

- Laser-induced incandescence is an appropriate tool for soot measurements inside engine combustion chambers and inside exhaust gases as well
- Applicable to production engines without any modification
- Simultaneous measurement of soot mass concentration and primary particle size ($\Rightarrow$ number concentration)
- High temporal resolution allows investigation of transient behaviour
- High sensitivity down to 100 µg/m^3 and even lower
- Robust technique with almost no interferences
- Potential as a standard method for maintenance and service