RESEARCH

# Applied Physics B Lasers and Optics



# Development of an optical particle number sensor based on continuous wave laser-induced incandescence

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#### Abstract

In this work, the development of a new particle counting sensor based on continuous-wave laser-induced incandescence is described. We demonstrate the sensor, which makes use of a focussed laser beam emitted by a compact laser diode, as an alternative device to established laboratory techniques to count soot particles. Simulations help understanding the cause–effect relationships based on known theory with the implementation of specific laser beam characteristics that influence the absorption process. The main goal of this work was to demonstrate the sensor's ability to determine the soot particle number concentration of a test aerosol. This has been confirmed through a comparison with established laboratory instruments, like scanning mobility particle sizer or condensation particle counter. The observed size dependency of the laser induced incandescence sensor's detection efficiency was successfully corrected with prior knowledge of the investigated particle size distribution. Furthermore, the sensor's detection limit with respect to the particle diameter was determined to be well below 100 nm with a dependency on the applied laser power.

# 1 Introduction

Environmental concerns and a growing awareness of the health hazards of soot particles are leading to stronger air quality regulations and an increasing demand for measurement techniques that are suitable for the precise determination of nanoparticle number concentrations [1-3]. In addition to the measurement of particle mass, knowledge of the total particle number of soot is crucial, especially for ultrafine particles with diameters measuring less than

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100 nm. High number concentrations of such particles pose health hazards, especially due to their detrimental effects on the respiratory system [4–6]. For these reasons, the development of reliable measurement techniques to count the number of ultrafine particles in diverse measurement environments is of high interest.

Commercial optical scattering-based sensors are limited to the detection of particles with diameters above approximately 100 nm [7]. Condensation particle counters (CPCs) are a type of optical particle sensors with an extended capability to measure particle sizes down to a few nanometres and are typically used in a laboratory environment to determine soot particle number concentrations [8]. To generate a measurable amount of scattered light, particles are inflated in size through the adsorption of a liquid. However, CPCs are bulky, costly and have the disadvantage of needing an additional fluid to serve as condensation nuclei (such as butanol, isopropanol, or water). To gather information about the particle size distribution, a CPC must be combined with an electrostatic or aerodynamic classifier to function as a scanning mobility particle sizer (SMPS) [9].

Sensors based on the principle of laser-induced incandescence (LII) can be seen as a versatile alternative since this optical method allows for an in situ investigation of various nanoparticle properties. The LII setups that are widely used in research of combustion-generated nanoparticles are normally based on the use of nanosecond pulsed laser sources [10, 11]. Typical applications are the measurement of soot volume fraction and mass concentration [12, 13]. The LII signal shape, or more precisely its decay time, yields an indication for the primary particle size, and under some assumption their size distribution, but individual particles are not distinguishable with these pulsed LII setups. Information on individual particle level can, however, be gathered with the commercially available SP2(-XR) LII sensor, which is using a continuous wave (CW) laser [14, 15].

Whereas most of the existing LII setups use bulky and expensive Nd:YAG lasers, this work aims to develop a sensor with a more compact design to be used in various mobile measurement environments. The goal for our sensor concept is the determination of the nanoparticle number concentration and the particle size distribution including particles with sizes below 100 nm. In an automotive environment, a measurement close to the particle source is necessary which requires a compact and costeffective sensor design. Using the principle of continuous wave laser induced incandescence, the sensor has functional similarities to the SP2 instrument mentioned before. However, there are also significant differences regarding the laser source, the general optical design and the interaction between the laser beam and the soot particles, which are discussed in more detail in the following sections.

# 2 Theoretical approach and simulation of the laser-induced incandescence process

To develop a functional sensor setup with the characteristics mentioned before, the following key design requirements must be considered: a sufficiently high laser power density for the particle excitation, the collection efficiency for the emitted LII light, and a high stray light rejection in the detection area. These main design aspects have been studied with the subsequent simulation approaches.

A suitable detector type must be chosen to deal with the low light intensities emitted by individual incandescent soot particles with a sufficient dynamic range to cope with the strongly varying peak signal values as a function of the particle's size. As an example, the total light intensity emitted by a 30 nm-diameter particle is in the range of tens of nanowatts, whereas a 200 nm-diameter particle emits a total intensity approximately three orders of magnitude higher, assuming a similar excitation temperature. Depending on the collection optics, specifically the usable solid angle, only a small percentage of the overall emitted LII light is collected.

#### 2.1 Implementation of LII models

The sensor model incorporates the energy transfer mechanisms describing the particle's incandescence and the necessary equations to calculate the intensity distribution of the laser light. The theoretical background of the main LII processes has been investigated since several decades and is discussed in detail in the literature [10, 11, 16]. The main contributions to the energy transfer between the particle, the laser, and the environment and, therefore, the temporal change of the internal energy  $U_{int}$  are heat exchange processes through absorption of the laser light  $Q_{abs}$ , cooling through conduction  $Q_{cond}$ , particle sublimation  $Q_{sub}$ , and to a smaller degree radiation of incandescence light  $Q_{III}$ :

$$\frac{\mathrm{d}U_{\mathrm{int}}}{\mathrm{d}t} = \dot{Q}_{\mathrm{abs}} + \dot{Q}_{\mathrm{cond}} + \dot{Q}_{\mathrm{sub}} + \dot{Q}_{\mathrm{LII}}.$$
(1)

Whereas the absorption term  $\dot{Q}_{abs}$  is positive, the loss terms for conductive cooling  $\dot{Q}_{cond}$ , sublimation  $\dot{Q}_{sub}$ , and radiation of LII light  $\dot{Q}_{LII}$  have negative values. The effects of oxidation, annealing, and thermionic emission are not considered in this equation due to their small contribution to the energy transfer compared to the other factors [11, 16, 17].

As a consequence of the optical design, specifically the use of excitation by a CW light source, the energy exchange processes of our approach mainly differ by the absorption term, i.e., LII excitation, relative to most other employed approaches presented in literature.

#### 2.2 Description of the laser-particle interaction

Laboratory setups based on pulsed LII and the commercially available Single Particle Soot Photometer (SP2, by Droplet Measurement Technologies) commonly use an Nd:YAG laser with either its fundamental wavelength of 1064 nm or harmonics like 532 nm.

To create a more robust and cost-effective sensor design with further potential for miniaturisation, the use of commercially available single mode laser diodes is investigated. The laser diode is operated in continuous wave mode instead of pulse mode. Due to a smaller measurement volume relative to most conventional approaches, individual particles can be counted. Therefore, the particle-laser interaction of this novel sensor principle approach is more comparable to SP2. For the SP2, particles are introduced into the cavity of an Nd:YAG laser with an intracavity power density of  $\approx 1 \frac{MW}{cm^2}$  [14]. In contrast to pulsed LII, the interaction time between the laser light and the particle is on the order of a few microseconds instead of nanoseconds. This difference in interaction time influences the relative contributions of the individual terms of the energy-balance equations. Bambha and Michelsen describe conductive cooling and absorptive heating to be the most dominant factors for CW LII [18].

In general, the absorbed energy rate is given by

$$\dot{Q}_{abs} = \sigma_{abs} \cdot E_e(t) \tag{2}$$

with  $\sigma_{abs}$  denoting the particle's absorption cross section and  $E_e(t)$  being the time-dependent irradiance. Whereas for pulsed LII systems  $E_e(t)$  is determined based on the laser fluence (in  $\frac{J}{m^2}$ ) and the pulse duration, this factor depends on the spatial intensity distribution and the particle velocity through the laser profile for the CW LII systems presented here. For pulsed LII, the rise time can be on the order of picoseconds to nanoseconds compared to microseconds for our setup.

To create an area with a high enough power density for a sufficient heating of the investigated particles with the limited output power of available laser diodes, the laser beam is focused to a small spot with only several micrometres in size. The shape of the focus spot is approximated by a Gaussian beam geometry [19]. The distribution of the optical energy within the focus area is calculated based on the properties of the optical components or by using measured values of the spot diameter and the Rayleigh length for the given experimental setup. With this, the time-dependent irradiance is calculated by

$$E_e(t) = I_0 \cdot \exp\left(\frac{-2 \cdot z(t)}{w_{0,\text{laser}}^2}\right).$$
(3)

Here,  $I_0$  is the maximal laser intensity, z(t) the time-dependent particle position perpendicular to the optical axis, and  $w_{0,\text{laser}}$  the laser beam diameter in the focus plane. This equation is valid for a perpendicular travel direction of the soot particles relative to the laser beam propagation.

The absorptive heating rate for a soot particle travelling through the detection area is then calculated by

$$\dot{Q}_{abs} = \sigma_{abs} \cdot I_0 \cdot \exp\left(\frac{-2 \cdot z(t)}{w_{0,laser}^2}\right).$$
(4)

In case of a parallel orientation, the equation changes due to a change of  $E_e(t)$  to

$$\dot{Q}_{abs} = \sigma_{abs} \cdot I_0 \cdot \frac{1}{\sqrt{1 + \frac{z(t)^2}{z_{R,x}^2}} \cdot \sqrt{1 + \frac{z(t)^2}{z_{R,y}^2}}}$$
(5)

with z(t) as the time-dependent particle position parallel to the optical axis and  $z_{R,x}$ ,  $z_{R,y}$  as the Rayleigh lengths in x- and y-direction, respectively.

# 2.3 Particle signal simulation with discrete time steps

Using the theoretical equations describing the LII mechanisms and the intensity distribution of the laser light, a soot particle travelling through the detection area of the sensor is simulated. The particles are modeled as isolated spherical particles with variable diameter  $d_{\rm P}$ , as for example discussed by Melton [20]. Their properties are further described by the thermal accommodation coefficient  $\alpha_T = 0.3$  [23], the particle density  $\rho_{\rm P} = 1860 \frac{kg}{m^3}$  [22], and the absorption function E(m) = 0.35 [21]. Deviations of the experimental results from the simulations must be considered due to this simplification.

Here, the particle travels with a constant velocity and a fixed angle with respect to the laser beam propagation. The exact travel trajectory can be varied to describe a statistical distribution of the travel paths within the detection area. For each discrete time step, the energy balance equation, the corresponding differential equations, and thus the particle's temperature T are calculated following the methodology in [16]:

$$\frac{\mathrm{d}T}{\mathrm{d}t} = \frac{1}{Mc_{\mathrm{S}}(T)}(\dot{Q}_{\mathrm{abs}} + \dot{Q}_{\mathrm{cond}} + \dot{Q}_{\mathrm{sub}} + \dot{Q}_{\mathrm{LII}}) \tag{6}$$

with the particle mass M and the temperature dependent specific heat of the solid particle  $c_{\rm S}(T)$ .

According to Liu et al. [24], the temporal change in particle diameter  $d_p$  due to sublimation is given by:

$$\frac{\mathrm{d}d_{\mathrm{P}}}{\mathrm{d}t} = \frac{2}{\rho_{\mathrm{P}} \cdot \pi \cdot d_{\mathrm{P}}^2} \cdot \frac{\mathrm{d}M}{\mathrm{d}t}.$$
(7)

Using these differential equations, the time-dependent particle temperature T(t), the particle diameter  $d_{\rm P}(t)$ , the emitted LII light, and its spectral distribution based on Planck's law can be calculated. The rate for the LII emission is given by

$$\dot{Q}_{\rm rad} = -\frac{198.97\pi^3 d_{\rm p}^3 (k_{\rm B}T)^5 E(m)}{h({\rm hc})^3}$$
(8)

with the Planck constant h, the Boltzmann constant  $k_B$  and the speed of light c.

With a consideration of the geometrical properties of the lenses in the detection path and the spectral efficiency of the filter configuration, the collected amount of LII radiation is determined. With further knowledge of the detector's properties, the expected electrical signal shape can be calculated.

Figures 1, 2, 3 and 4 present results of respective simulations for a 100 nm particle, traveling with a velocity of  $1\frac{\text{m}}{\text{s}}$ , heated by a laser with a wavelength of 800 nm and an optical output power of 200 mW in the parallel configuration. The change in particle temperature, its diameter, the



Fig. 1 Simulation result for the time-dependent particle temperature of a 100 nm particle heated by a laser with an optical output power of 200 mW. The temperature reaches a maximum value of nearly 3500 K



Fig. 2 Simulation of the time-dependent behaviour of the particle diameter for a 100 nm particle heated by a laser with an optical output power of 200 mW

dominating energy fluxes, and the captured LII signal are shown as functions of travel time. For all profiles, an abrupt change with high gradient can be observed, since initially the particles approach the laser focus spot through an area of very low laser fluence and thus are heated slowly. The strong focussing of the laser beam is associated with a very steep increase of the fluence with the corresponding heating. After the focus, the particles are rapidly entering a region with significantly lower fluence.

The particle temperature reaches a maximum of nearly 3500 K (Fig. 1). The slightly asymmetrical shape of the temperature profile can be explained by the size reduction due



Energy Flux (W)

10

Fig. 3 Dominating heat fluxes for a 100 nm particle heated by a laser with an optical output power of 200 mW

1.0

Time (ms)

1.5

2.0

0.5



**Fig. 4** Collected LII signal incident on the detector for a 100 nm particle heated by a laser with an optical output power of 200 mW

to partial sublimation. During the incandescence process, the particle diameter decreases to 20 nm (Fig. 2). Figure 3 shows the temporal profile of the main energy fluxes. The great majority of the absorbed energy is conducted through the surrounding gas, here with a temperature of 330 K. To a much smaller degree, energy is exchanged through sublimation and LII radiation. The collected LII signal incident on the detector is shown in Fig. 4.

# 3 LII sensor design

This section describes the considerations leading to the design of the LII sensor and the choice of suitable components for its realisation. Figure 5 shows a schematic representation of the sensor setup including the optical components





Fig.6 Experimental setup of the CW-LII sensor highlighting the main components. The demonstrator's dimensions are roughly  $30 \times 45 \times 10$  cm

and the corresponding laser and LII light paths. A photograph of the experimental setup is displayed in Fig. 6. The laser module consists of a laser diode with a wavelength of 830 nm and an aspherical lens with a focus length of 15 mm to collimate the divergent laser light (L1). The lenses in front of the soot inlet (L2) and in front of the detector (L3) have focus lengths of 25 and 35 mm, respectively.

To optimise the sensor dimensions, the sensor setup is designed with partially overlapping excitation and detection paths. For the excitation of soot particles, the laser beam is shaped with collimation and focus lenses to create a well-defined focus area within the measurement pipe. The "particle lens" (L2) in Fig. 5 is used to focus the laser light as well as to collect the emitted LII light. The LII light of appropriate wavelengths then passes through the dichroic mirror and an additional filter system before it is projected onto a detector. The main advantage of this confocal setup are the requirement for only one single optical access to the measurement area, compared to three for the SP2-XR or five for the SP2, and a significant reduction of the complexity of the optical alignment.

Commercially available laser diodes are chosen as laser source to enable a robust and cost-effective sensor with the potential for miniaturisation. A crucial issue for the sensor functionality is the creation of a region with sufficiently high power density for the desired particle heating process. High-quality lenses are necessary to shape the laser beam with minimal aberrations. Edge-emitting laser diodes emit divergent light with direction-dependent divergence angles due to the rectangular shape of their emission facet. The diverging laser beam is collimated by a plano-convex lens. For a compact realisation of the sensor design, the now parallelised laser beam with an elliptical shape and semi-axes of several millimetres is reflected by a shortpass dichroic mirror. Subsequently, a plano-convex spherical (or aspherical) lens focuses the laser beam onto a fine elliptical spot with semi-axes of a few micrometres to generate a high power density within the focus spot area. To separate the optical system from the aerosol flow in the soot pipe, an additional glass window is positioned behind the lens. On the other side of the soot pipe, an anti-reflection coated glass window is followed by a beam trap to absorb most of the transmitted laser light and to prevent it from re-entering into the system.

The particles travel perpendicular to the laser beam through a soot inlet pipe with an inner diameter of several millimetres. Assuming a homogeneous distribution of the particles within the measurement chamber's cross section, only a small fraction of the particles travels through the detection area with a high laser power density in the laser beam's focus area. The particles absorb optical energy and are heated to high temperatures above 3000 K. These particles emit incandescence light into all spatial directions. A small fraction (about 3% for the implemented setup) is collected by the focus lens and collimated.

The collinear design between the dichroic mirror and the measurement pipe allows for a compact sensor design with the possibility of miniaturisation. After passing through the shortpass dichroic mirror and the edgepass filters, the LII light is focused onto the detector, where a photo current is induced.

The detector is chosen to be a silicon photomultiplier (SiPM) array since this compact detector type is sensitive to low light intensities. It is operated in the so-called Geiger mode so that in each of the photodiodes one photon can trigger a charge carrier avalanche. With its array structure and several hundred individual pixels that are connected in parallel, a sum signal with a high dynamic range is produced. An electrical circuit transforms and amplifies the signal to a measurable voltage, which is recorded with an oscilloscope (Tektronix DPO 7054C) or a data acquisition card (Spektrum M2p5962-x4j with a sampling rate of 125 MS/s).

## 3.1 Choice of the optical components

The optical components with the highest influence on the performance of the LII sensor are the laser diode, the collimation and focus lenses to shape the laser beam and a filter system that suppresses stray light in the detection area.

For the LII process, it is crucial to choose a suitable laser source regarding its wavelength, optical output power, and beam quality. Based on simulation results, an area with a laser power density between several 100 kW/cm<sup>2</sup> and 1 MW/cm<sup>2</sup> is required, which is similar to the values achieved with the commercial SP2 sensor.

Soot absorbs light in a broad spectral range from the ultraviolet to the mid-infrared, but depending on the laser wavelength, the particle's absorption cross section changes with

$$\sigma_{\rm abs} = \frac{\pi d_{\rm p}^3 E(m)}{\lambda_{\rm L}},\tag{9}$$

where  $\lambda_{\rm L}$  represents the laser wavelength [11].

A longer wavelength requires a higher laser power due to a decreasing absorption cross section of the particles and typically leads to larger focus areas of the laser beam. The theoretically achievable focus spot radius is limited by

$$w_0 \approx \frac{\lambda_{\rm L} \cdot f}{\pi \cdot R_0} \tag{10}$$

with the focus length f and the initial beam radius after collimation  $R_0$  [19].

Since achievable focus spot sizes with multi-mode lasers are significantly larger, higher optical output powers are necessary in this case, which result in a higher amount of stray light, an increased heat generation, and a more expensive electronic driver.

For this reason, a single-mode laser diode with an optical output power of 100 - 1000 mW is chosen to create small focus spots with diameters of about 10 µm for the required power density.

The LII signal from the heated particles has a broadband spectrum with light mostly in the visible into the IR range depending on the particle's maximum temperature. For a spectral distribution of the emitted LII light, the model relies on known LII equations [11]: To collect most of this signal with a given filter setup, it is advantageous to select a laser wavelength in the lower visible regime (< 500 nm) or in the near-infrared (> 800 nm). This allows for an efficient blocking of the laser light in the detector region while transmitting most of the LII signal to enhance the sensor's signal-to-noise ratio. However, using a laser in the lower wavelength regime can induce fluorescence light in the sensor setup that interferes with the desired LII signal and thereby reduces the signal-to-noise ratio of the detector [11].

The previously described simulation environment was used for the identification of a suitable laser source. With a parameter variation of laser power and laser wavelength, maximum particle temperatures and the amount of emitted LII signal were evaluated. The results led to the choice of a laser diode with a wavelength of 830 nm and an optical output power of 650 mW.

The optical elements are chosen to match the properties of the emitted laser light, like the laser diode's divergence angles (typically between 5° and 20°). The focus length of the laser collimation lens (L1) defines the diameter of the collimated light beam. A wider beam allows to focus the light onto a smaller spot, see Eq. 10. Apart from the lenses' focus lengths, the wavelength  $\lambda_{\rm L}$  further influences the focussing characteristics of the light beam. A plano-convex aspherical lens is chosen as collimation lens to minimise spherical aberrations and to improve the beam collimation. In this sensor setup, L1 has a focus length of 11 mm and collimates the divergent laser light (7° and 14°) to an elliptical beam with 0.7 mm and 1.4 mm semi-axes radii.

The shorter the focus length f of the focussing particle lens (L2), the smaller the focus diameter  $w_0$  can become. Since L2 also collects the LII signal, a lens with a high numerical aperture should be chosen to collect a maximum amount of the LII light, which is emitted in all spatial directions. Considering the geometry of the soot pipe, a lens with a diameter of 25.4 mm and a focus length of 30 mm is installed.

The spectral part of the collected and collimated LII light with wavelengths below 800 nm passes the shortpass dichroic mirror. To ensure the most efficient blocking of laser light in the detector path, additional shortpass filters are used. The setup includes a combination of reflective and absorptive filters, blocking wavelengths above 700 nm. This choice is a trade-off between the loss of detectable LII signal and an efficient blocking of the laser light.

The position of the detector is chosen to be beyond the focus plane of the detector lens (L3, d = 25.4 mm, f = 35 mm) to achieve a homogeneous illumination of the photomultiplier area and to avoid saturation of an unnecessarily small number of detector pixels in the centre. This enables a larger dynamic range of the output signal for a given detector architecture. The implemented SiPM detector consists of 667 pixels with pitches of 50  $\mu$ m, a total detection area of 1.3  $\times$  1.3 mm, and a maximum photon detection efficiency of 40%.

## 3.2 Construction of the sensor setup

The optical components are positioned within a 30 mm cage system (Thorlabs) to enable an accurate alignment. Depending on the needs, rotations and translations can be performed precisely. Furthermore, the possibility to easily change single components helps testing different variations. The sensor setup is shown in Fig. 6. The light paths are aligned with the use of beam characterizing instruments such as a scanning slit beam profiler or an optical power meter.

# 4 Laboratory setup

As soot particle source, a three-flame "miniCAST Real Soot Generator" by Jing Aerosol AG with propane as gaseous fuel is installed. The particle size distributions can be altered reproducibly by a change of the ratio between propane, oxygen, and nitrogen [25]. For the LII measurements performed in this work, particle distributions with mean diameters between 60 and 100 nm are generated (see SMPS measurements in Fig. 7). The generated distributions typically have a log-normal shape and contain particle sizes in the range from approximately 20– 200 nm. It has to be noted that due to its measurement principle, the SMPS measures the particle number concentration as a function of the electrical mobility diameter  $D_m$  [26]. Therefore, the particle diameters in the displayed distribution curves are the measured



**Fig.7** miniCAST particle distributions measured with a scanning mobility particle sizer. The particle distributions show comparable shapes with mean electrical mobility diameters of 92 nm, 73 nm and 60 nm

mobility diameters. However, LII models rely on the geometrical particle diameter. Nevertheless, the SMPS assures that the LII sensor is fed with a sample of known particle distribution. The sample air containing the soot particles is diluted by a Dekati eDiluter and two Dekati DI-1000 diluters. The eDiluter can be set to ratios between 1:25 and 1:125. The DI-1000 diluters exhibit dilution ratios of 1:8 per stage. Subsequently, it is passed through the LII sensor setup and to an SMPS. The pump of the SMPS is used to draw the aerosol through the LII sensor's measurement pipe. Simultaneously, an independent value for the total particle number concentration is generated with a separate CPC connected in parallel to the LII sensor behind the dilution stage. The reference measurement values are used to determine the sensor's sensitivity and size detection limit. Figure 8 schematically shows this measurement configuration. Since only a small fraction of all particles, that flow through the measurement pipe, is traversing the high-intensity focus area and excited to sublimation, the overall size distribution is not altered significantly by the LII sensor. Reference measurements with a switched-off laser confirm this.

# 5 Results and discussion

#### 5.1 LII event detection in output signal

For the data evaluation of the LII sensor signals, an oscilloscope (Tektronix DPO 7054C) with a time resolution setting of 5 ns is chosen. To detect LII signal events in the detector output trace, a trigger scheme with a voltagebased and duration-based threshold is used (see Fig. 9 for an example). The duration-based threshold is required to filter out short noise events with durations of a few hundred nanoseconds and relatively high maximum voltages. These noise events are the result of cross-talk between adjacent pixels within the detector. Since the duration of LII signals is expected to be on the order of several microseconds,



**Fig. 8** The concentration of a selected particle size distribution is reduced by diluters with varying dilution ratios and subsequently measured by a CPC, the LII sensor and an SMPS

Voltage (mV)

Fig. 9 Depiction of a typical LII event recorded by an oscilloscope. Noise events that surpass the voltage threshold are not registered because they don't exceed the required minimum time duration. The oscilloscope triggered to the rising edge of a main peak (visible at around 40 µs) with a minimum peak width of 2 µs

they remain largely unaffected by the additional temporal threshold. A purely voltage-based threshold would have to be set too high to prevent a triggering to these noise events. Thus, the sensor would not detect signals created by small soot particles, because they only reach similar voltage levels as induced by the system's noise. Since the LII signal durations are expected to be within the range of several microseconds, the additional temporal trigger threshold serves to distinguish short noise from longer LII events. In combination with a minimum time interval. a significantly lower voltage threshold can be set without triggering to noise events. The optimal threshold value depends on the setup's noise level. It is adjusted based on measurements with filtered air and increased until noise is not mistakenly detected as a particle event. For the present measurements, the voltage threshold value is set to 15 mV with a minimum time interval of  $2 \mu s$ .

The LII signal trace on the detector depends on numerous factors. The particle size directly influences the total signal intensity and the maximum value whereas the signal width is mainly a function of the particle speed. Exemplary signals are shown in Figs. 9, 10 and 11. Furthermore, the overall morphology of the soot particles and their aggregates influence the shape of the falling edges of the LII signal curves [18]. Figure 11 gives an example for the difficulty of detecting very small particle signals with the applied threshold method. An additional accumulation of pulses is visible towards the end of the recording time that cannot be clearly distinguished from noise and is not considered as a separated LII signal event.

Fig. 10 Two distinct LII events stemming from two separate particles are displayed. The signal shapes in this example differ from the signal given in Fig. 9, potentially due to a different particle geometry

# 5.2 Determination of the sensor performance

To judge the LII sensor's performance, its lower size detection limit is an important parameter. The goal of this work is to measure particles with diameters well below 100 nm. For soot measurement devices, the direct determination of a detection limit with a high accuracy poses difficulties since a test gas containing soot particles with ideally just a single, discrete size would be required. Furthermore, the performance of a size measurement on individual particle level with the available reference laboratory equipment is not feasible.

400

300

200



mulation of pulses is visible, that cannot be clearly distinguished from noise by the threshold method and is therefore omitted





Therefore, two different methods for the sensor characterisation were employed and compared. The first indirect procedure has the advantage of a shorter measurement time and lower effort and allows for the efficient study of causeeffect relationships and experiments with wider parameter variation. It is based on a ratiometric calculation method detailed below. The alternative method employs narrow sizeselected distributions produced using an additional classifier device (such as a Differential Mobility Analyzer (DMA) or an Aerodynamic Aerosol Classifier (AAC)). Results gained with this method enable the calculation of the sensor's sizedependent detection efficiency curve.

#### 5.2.1 Use of the indirect ratiometric method

The indirect procedure to determine the sensor performance consists of the following steps: The size distribution of particles created with a miniCAST soot generator is measured with an SMPS. Simultaneously, LII events registered with the laboratory LII sensor are recorded to obtain a total number of LII events per unit time with the threshold method described above.

Subsequently, an artificial detection limit is introduced for the size distributions measured with the SMPS (Fig. 12). This artificial limit is used to trim the SMPS data to mimic the higher detection limit of the LII sensor with respect to size.

The particle number concentration is then calculated by an integration of the measured particle number concentrations with  $d_{\rm P} > d_{\rm L,a}$ , with the artificial size limit  $d_{\rm L,a}$ . The artificial size limit is varied and the particle number concentration for larger particle diameters is calculated for each step. These calculations are performed for different particle size distributions (Fig. 13).

To reduce the influence of differences in sensitivities between the LII setup and the SMPS, ratios of these total particle counts for different particle distributions are calculated.

For the SMPS data this means for example:

Ratio of count rates (SMPS) = 
$$\frac{C_1(d_{L,a})}{C_2(d_{L,a})}$$
 (11)

for two particle distributions with the number concentrations  $C_1$  and  $C_2$  for all particles sizes with  $d_P > d_{L,a}$ .

The ratio is also calculated for the time-integrated number of registered LII events per unit time, which are counted for the respective size distributions:

Ratio of count rates (LII) = 
$$\frac{L_1(d_{\text{L},a})}{L_2(d_{\text{L},a})}$$
 (12)

for two particle distributions with the LII count rates  $L_1$  and  $L_2$ . For each measurement sequence, this gives a constant value.

The intersection between the corresponding SMPS and LII ratio curves is calculated and serves as a figure of merit to provide information about the performance of the LII sensor, see Fig. 14. For the particle size corresponding to this intersection, the count ratio for the LII events registered for the two investigated size distributions equals the ratio of the particle number concentration for diameters larger than this value.



**Fig. 12** A variable artificial detection limit for the SMPS data is introduced (see vertical line). Starting from this mobility diameter value, the total particle concentrations are integrated



**Fig. 13** The integrated particle number concentrations starting at the variable artificial detection limits is displayed. When starting the integration at small diameters  $d_{L,a}$ , the total number concentration is higher for size distribution 2. This changes when increasing the lower limit, since distribution 1 consists of a higher number of large particles



**Fig. 14** The intersection between the curve for the ratio between integrated particle number concentrations of two different distributions and the corresponding LII counts gives a figure of merit for the sensor's performance. For this example, the figure of merit is 130 nm

The advantage of this method is a fast and easy way to compare between different setups and parameter variations like the laser power or the particle velocities within the detection area. Since the size distributions are not pre-selected, the high number concentrations allow for short measurement times.

For the exemplary setup with an NIR laser with  $\lambda = 830$  nm and P = 650 mW, the intersection of the ratio lines is at 130 nm.

It must be noted that the size-dependent detection efficiency of the LII sensor is not considered for these calculations.

For a first approximation of the sensor's detection limit, an additional DMA is implemented to filter the particle size distributions emitted by the soot generator. Due to its working principle, distributions with a main peak at the chosen diameter and side peaks are created [26]. The main peak corresponds to singly charged particles, whereas the side peak corresponds to double-charged particles. The side peak structure is presented in another context later in this work, see Fig. 16. For one sensor realisation, with a setting of the DMA-selected main peak at 20 nm, no events are registered by the LII sensor. Since the side peak contains particle diameters of up to 33 nm, a detection limit above this value can be inferred. With a DMA setting of 30 nm, LII events could be recorded successfully, which are attributed to the side peak at 58 nm. Based on these measurements, the lower detection limit of this sensor setup lies in the size range between 33 nm and 58 nm.



**Fig. 15** The figure of merit determined with the ratiometric method shows an initial strong improvement with increasing laser power before the positive effect diminishes for values above 650 mW



**Fig. 16** The particle size distribution is narrowed down with the use a DMA. Particles with a narrow main peak diameter and a side peak diameter are transmitted due to the work principle of the apparatus

#### 5.2.2 Variation of the laser power

Since a change of the laser power directly influences the time-dependent irradiance, particles with different diameters will be affected differently. Smaller particles possess a larger surface-to-volume ratio and thus an increased cooling rate. To investigate the influence of the laser power on the sensor performance, six different laser currents are applied, yielding a variation in laser output power between 250 mW and 700 mW. The number of registered LII counts increases for higher laser power settings, as expected. The count ratio between the LII counts measured for the two miniCAST operation points increases simultaneously, as the

SMPS curve remains constant. The intersection between the SMPS and LII curves shifts to smaller particle diameters with increased laser power, as displayed in Fig. 15, indicating a better performance of the sensor for detecting particles of small sizes.

By plotting this figure of merit against the corresponding laser power, a non-linear behaviour is observed. For higher power densities, the improvement potential decreases and the figure of merit shows an asymptotic behaviour above approximately 650 mW such that a further increase of laser power does not improve the sensor's sensitivity. This behaviour implies that the sensor's performance can be improved by creating a detection area with higher power densities. However, this works only up until a certain point. Possible reasons for the saturation are an increase of unwanted laser stray light and a saturation LII signal output.

Further optimisation beyond the current experimental setup, i.e. through an increase of the optical aperture within the geometric constraints of the system and spectral filtering shall be tackled in future work. Another promising approach is an improvement of the detection method for LII events by implementing more sophisticated algorithms, e.g., with methods of machine learning, to distinguish weak signals corresponding to small particles from noise.

#### 5.2.3 Determination of the detection efficiency

Whereas the previously described method provides a fast way to compare different setup versions, a more precise method is needed for the evaluation of the sensor's detection efficiency. The size-dependent detection efficiency curve is calculated with the data from size selected particle measurements. For a narrower size distribution of the particles, a DMA is placed in front of the LII sensor to select and separate a specific particle diameter. The working principle of a DMA results in a selection of particles with a certain mobility diameter [26]. Therefore, larger particles with the same mobility as the desired particle size will also pass the setup leading to side peaks in the distribution. As an example, Fig. 16 shows the SMPS-measured distributions for two different DMA settings.

The side peaks lead to some level of imprecision in the measurement of the detection efficiency, since the signal of a smaller particle travelling through the center of the focus area can be comparable to the signal of a larger particle traveling at the edge, where it absorbs less laser energy and thus emits less incandescence light, as displayed in Fig. 17.

Nevertheless, the results are significantly more accurate compared to measurements with the broad original particle size distributions. To further improve the measurement, the DMA could be replaced or combined with an AAC, which does not exhibit side peaks in its particle distribution. This is planned for future experiments.



**Fig. 17** Depending on the particle size, the detection area for a sufficient particle heating changes. Whereas smaller particles need to pass through the areas with the highest power density, larger particles can emit detectable amounts of LII signal when passing further away from the focus spot centre. The region with the highest laser intensity is coloured red, lower intensities are displayed in blue. The arrows display possible particle trajectories



**Fig. 18** The normalised sensitivity given by the LII detection rate divided by the measured particle concentration (with an SMPS) is shown as a function of the selected particle mobility diameter. Especially, the measurement with 400 mW laser power shows a saturation of the efficiency for particles larger than 140 nm. The horizontal error bars show the standard deviation of the mean diameters compared to the selected particle sizes due to the side peaks in the distributions, as displayed in Fig. 16

For a large range of selected particle diameters, LII counts were registered. Normalised sensitivity values were calculated by dividing the LII count rates by the total particle number concentration from SMPS measurements. Figure 18 shows the sensitivity curves for three different laser power values.

All three curves display a decrease of the slope for larger particle sizes. Especially for the setting of 400 mW, a clear saturation of the sensitivity with an increase of the particle diameters is visible. This indicates that the detection efficiency reaches a maximum value for particles larger than approximately 140 nm. As the sensor is based on the interaction between particles and laser light within the focus area of a near-Gaussian laser beam, there is no large-scale region with a constant, homogeneous energy or power density within the measurement chamber. Larger particles can absorb light more efficiently and thus can travel through a wider region around the laser focus whilst still emitting a measurable LII signal. Smaller particles require higher excitation energy levels, which are present close to the focus point, see Fig. 17. Above a certain size value, the detection area becomes nearly constant, leading to the observed constant sensitivity to detect these particles.

#### 5.3 Particle number concentration measurement

The key measurement function of the LII sensor is the counting of individual nanoparticles. Particle counting devices like CPCs are commonly used to measure particle number concentrations in the sub-100 nm range. Since they are too bulky and costly for many mobile applications and require an additional liquid, a reliable counting function of the LII sensor enables a wide range of additional applications. For the following evaluation, a CPC (Airmodus Ltd.) and a SMPS (TSI GmbH) are used as reference devices for comparison with the LII measurements.

For the investigation of the LII sensor's particle counting capability, the measurement environment shown in Fig. 8 with a variable dilution stage is used. The SMPS measurements presented in Fig. 19 show that by modifying the dilution ratio, the particle size distribution and its mean diameter are not changed but only the total concentration is varied.

As an example, it can be seen in Fig. 19 for a dilution ratio (DR) of 25 (solid curve), the concentration is 1.6 times higher compared to a DR of 40 (dashed curve).

In Fig. 20, the number of LII events per minute is compared to the particle number concentration measured with the CPC for different size distributions. The distributions' mean diameters vary between 60 nm and 92 nm. Each curve shows the expected linear dependency between detected LII events and the total particle number concentration. The total particle number concentration can be deduced from the LII count frequency using the regression in Fig. 20 as calibration curves. However, it can also be observed that the slopes clearly differ for each investigated size distribution. For larger mean particle sizes, the regression line slope becomes steeper compared to the distributions with smaller particles.

These observations can be explained by the particle sizedependency of the sensor's detection efficiency. To accurately determine the particle number concentration independently of its size distribution, a suitable correction of this size dependency has to be implemented. The correction process is based on the knowledge of the size distribution. In this work, the size data recorded by an SMPS is used.

For independent and mobile measurement applications, where such data is typically not available, the goal is to derive the required size information directly from the LII signal shapes. Since the emission rate of the LII signal is a function of the particle size (see Eq. (8)), the registered signal shapes could be used for the estimation of the particle diameters, as will be evaluated in future work.

Using the mean diameters of the distributions as an input parameter, the weighted LII counts are calculated



 $\begin{array}{c} \text{Size Distribution 1} \\ \text{Size Distribution 2} \\ \text{Size Distribution 3} \\ \text{Size Distrib$ 

**Fig. 19** A change of the dilution ratio (DR) of a Dekati eDiluter influences the total particle concentration but not its mean particle diameter or size distribution. The distributions were recorded with an SMPS

**Fig. 20** Comparison of detected LII events per minute to particle concentrations measured by a reference CPC. The linear behaviour for each investigated size distribution confirms the capabilities of the LII sensor to count soot particles. The difference in slope values can be explained by the particle size dependence of the sensor's detection efficiency



**Fig. 21** After correcting the LII counts using the mean particle diameter for each distribution, a size-independent correlation with the particle concentration can be observed



Fig. 22 The estimated concentration values resulting from the LII counts adjusted with the calibration and size correction are in good agreement with the reference concentration values measured with a CPC with a relative error of less than 18%

by dividing the measured LII counts by the corresponding mean diameters to the fourth power. The weighted values are presented in Fig. 21. The linearity between corrected LII event counts and the particle number concentration is now independent of the investigated size distribution and shows a linear behaviour for all measurement points.

By combining the previously determined regression lines and the calculation of the estimated number concentration with these weighted results, the sensor can give a good estimation of the number concentration, see Fig. 22. A relative error of less than 18% compared to the reference CPC data is achieved for this measurement campaign. This error value is the highest deviation of the measured values from the reference concentrations.

# 6 Conclusion

In this work, we demonstrate a continuous wave LII sensor as an alternative device to established laboratory techniques to count soot particles. We developed a simulation model for a CW LII device that makes use of a focussed laser beam emitted by a compact laser diode and built up an experimental setup guided by its results. With the measurement results for narrow size distributions, its detection limit with respect to the particle diameter was determined to be well below 100 nm. We investigated the dependence of the sensor performance on the laser power. The determination of the optimal power value of the used sensor is crucial to find a balance between a sufficient excitation of small particles and the detrimental effect of a higher noise level due to laser stray light. The optimal laser power, considering the cost-benefit factor, is dependent on the optical design and was found to be around 650 mW for our sensor setup.

The main goal of this work was to demonstrate the sensor's ability to determine the soot particle number concentration of a test aerosol. This has been confirmed through a comparison with established optical laboratory instruments, i.e., SMPS or CPC. The observed size-dependency of the LII sensor's detection efficiency was successfully corrected with prior knowledge of the investigated particle size distribution. In this work, data from reference SMPS measurements was used for this purpose.

Future work will include the improvement of the signal evaluation procedure to optimise the sensor's performance and the detection of particle diameters below 50 nm. Measurements in combination with size selection by an Aerodynamic Aerosol Classifier can increase the accuracy of the determination of detection efficiency and detection limit. In addition, the derivation of particle size distribution and geometry based on the LII signal shape will be investigated and implemented. Using this size information, an online calculation of the correction for the sizedependent efficiency and an accurate value for the particle number concentration can be achieved.

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#### **Declarations**

Conflict of interest The authors declare no competing interests.

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