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Agenda

Sunday June 10th

17:00 – 18:00	Registration
18:00 – 21:00	Dinner and get together

Monday June 11th

08:15 – 08:40	Registration
08:40 – 09:00	Welcome and introduction
09:00 – 10:00	Oral session 1: Soot Properties
09:00	A new approach for in situ soot size distribution measurement based on spectrally resolved light scattering ***M. Bouvier****, J. Yon, G. Lefevre, F. Grisch*
09:20	Insights on laser-baked soot ***F. Migliorini****, S. De Iuliis, R. Dondè, M. Commodo, P. Minutolo, A. D’Anna, L. Ferrero*
09:40	Raman spectroscopy on soot produced from a mini-cast soot generator: impact on structure from heating in air and nitrogen up to 900°C ***K. C. Le****, S. Török, T. Pino, P.-E. Bengtsson*
10:00 – 10:20	Discussion about future interlab comparison (to be continued during poster session 2) Raphael Mansmann
10:20 – 10:50	Coffee break
10:50 – 12:00	Oral session 2: Technique Extensions & LII Process Details
10:50	LII and MAE measurements in a laminar diffusion flame to assess the ISF database consistency *B. Franzelli,* ***M. Roussillo*** *, P. Scouflaire, J. Bonnety, R. Jalain, T. Dormieux, S. Candel, G. Legros*
11:10	Coupling of cavity-ring-down extinction and laser induced incandescence to determine soot volume fractions in a nucleation and a sooting premixed flames ***P. Desgroux****, C. Betrancourt, X. Mercier, F. Liu*

11:30	LII particle-size imaging with an ultra-high-speed CMOS camera ***E. Cenker****, S. Skeen, Y. Chen, D. Richardson, C. R. Shaddix, D. R. Guildenbecher*
11:45	Turbulent flame LII particle sizing via ultra-high-speed imaging ***Y. Chen****, D. R. Richardson, E. Cenker, B. R. Halls, S. Skeen, C. Shaddix, D. R. Guildenbecher*
12:00 – 13:20	Lunch
13:20 – 14:40	Discussion session 1
13:20	Discussion 1: Determination of key parameters for LII Per-Erik Bengtsson & Eric Therssen
14:00	Discussion 2: Supplementary and combined techniques Emre Cenker & Jérôme Yon
14:40 – 15:10	Coffee break
15:10 – 16:30	Oral session 3: New Technical Approaches & Applications I
15:10	Thermographic and two-phase PIV based on LII signal from submicron black particle tracers *L. Fan, D. McGrath, H. Zhong,* ***S. Hochgreb***
15:30	Laser induced incandescence (LII) using a long-pulsed fibre laser for in-situ study of soot in flames *R. Roy, Gordon Humphries,2, J.D. Black,* ***I.S. Burns***
15:50	Two-dimensional LII for in-situ soot characterization of propane flames and influence of additives in a 100 kW oxy-fuel furnace *J. Simonsson,* ***M. Mannazhi****, A. Gunnarsson, D. Bäckström, K. Andersson, P.-E. Bengtsson*
16:10	Investigation of soot formation in a novel diesel fuel burner ***N. Palazzo****, M. Kögl, L. Zigan, F.J.T. Huber, S. Will*
16:30 – 18:00	Free afternoon
18:00 – 19:30	Dinner
19:30 – 23:00	Poster session 1
19:30 – 20:30	Advisory committee closed meeting

Tuesday June 12th

09:00 – 10:00	Oral session 4: Modelling & Evaluation
09:00	A calibration strategy for planar laser-induced incandescence measurements at increased pressure *R. Hadef,* ***K.P. Geigle***
09:20	Effect of detection wavelengths on soot volume fraction measurements using auto-compensating (two-color) LII ***F. Liu***, *G.J. Smallwood*
09:40	Can soot primary particle size distributions be determined using laser-induced incandescence? ***F.J. Bauer***, *K.J. Daun, F.J.T. Huber, S. Will*
10:00 – 10:30	Coffee break
10:30 – 11:50	Discussion session 2
10:30	Discussion 3: Pulsed and CW LII: modeling, evaluation and unresolved questions Kyle Daun & Raphael Mansmann & Joel Corbin
11:10	Discussion 4: Non-soot LII Christoph Schulz
12:00 – 13:30	Lunch
13:30 – 14:30	Oral session 5: LII on Engineered Particles
13:30	Predicting the heat of vaporization of iron at high temperatures using TiRe-LII and Bayesian model selection ***T. A. Sipkens***, *S. J. Grauer, P. J. Hadwin, K. J. Daun*
13:50	Transition from laser-induced incandescence (LII) to laser-induced breakdown spectroscopy (LIBS) on elemental nanoparticles **J. Menser**, K.J. Daun, T. Dreier, C. Schulz
14:10	Laser-induced incandescence on metallic nanoparticles: Investigating effect of plasma thermal bremsstrahlung emission on peak temperature pyrometry inference ***S. Talebi Moghaddam***, *K. Daun*
14:30 – 15:00	Coffee break
15:15 – 18:00	Trip to Monastery Andechs with guided tour
17:30 – 20:00	Banquet / Dinner (Andechs)
20:30 – 23:00	Poster session 2

Wednesday June 13th

09:00 – 09:40	Discussion session 3
09:00	Discussion 5: Emitted and ambient aerosols Greg Smallwood & Francesca Migliorini
09:40 – 10:10	Coffee break
10:10 – 11:30	Oral session 6: Emitted and ambient aerosols / Applications II
10:10	SP2-XR: The Next Generation of Single Particle Black Carbon Instruments ***A. Attwood***, *H. Schulz, G. Granger, M. Zanatta, A.B. Herber, D. Baker and R. Gerdes*
10:30	Method and application of ambient black carbon mixing state measurements with the Single Particle Soot Photometer (SP2) ***R. L. Modini***, *J. Yuan, M. Zanatta, T. Müller, B. Wehner, and M. Gysel*
10:50	Laser-based experimental investigation on soot evolution during coal combustion in O_2/N_2 and O_2/CO_2 conditions ***J. Wu***, *L. Chen, P.-E. Bengtsson, J. Zhou, J. Zhang, X. Wu, K. Cen*
11:10 – 11:30	Discussion about future interlab comparison, wrap up (Mansmann/Daun)
11:30 – 12:00	Summary and closing remarks
12:00 – 13:30	Lunch and farewell, optional travel to Erlangen or Stuttgart
18:00 – 20:00	Optional dinner in either Erlangen or Stuttgart (Depends on choice of labtour)

Thursday June 14th

09:00 – 12:00	Optional labtour at the Institute of Engineering Thermodynamics in Erlangen or the German Aerospace Center in Stuttgart.

Posters

1	Muhammad Asif	Determination of optical properties of non-soot nanoparticles generated by microwave plasma via line-of-sight attenuation (LOSA)
2	Simon Aßmann	Investigation of soot formation in 2D by combined multi-angle light scattering and laser-induced incandescence employing a shielding approach
3	Emre Cenker	Effects of soot volume fraction on bath-gas heating and particle sizing during LII
4	Joel Corbin	Laser-induced incandescence of aircraft engine black carbon: sensitivity to laser fluence
5	Gordon Humphries	Photoacoustic Measurement of Soot in a Flat Flame with a High Rep Rate Fibre Laser
6	Niklas Jüngst	Multi-diagnostic imaging of evaporating fuel wall-films in combustion as a source of PAH and soot
7	Fengshan Liu	The morphology of soot aggregates generated in ethylene and propane inverse diffusion flames at different oxygen indexes
8	Raphael Mansmann	LIISim: A modular signal processing toolbox for laser-induced incandescence measurements
9	Stanislav Musikhin	Temporally- and spectrally-resolved LII measurements on a standard flame using a streak-camera and multichannel PMT setup
10	Manu Naduvil Mannazhi	Laser diagnostics for soot for high pressure CH4-air diffusion flames
11	Mathieu Roussillo	LII measurements in a Confined Swirled Sooting Flame under Perfectly Premixed Rich Conditions
12	Robert Roy	Laser induced incandescence imaging in diffusion flames of liquid fuels relevant to biomass combustion
13	Timothy Sipkens	What is hiding in the intensity scaling factor and what can be gained from analyzing its temporal variation?
14	Sina Talebi Moghaddam	Neutral bremsstrahlung emission in laser-induced incandescence experiments on soot and silver nanoparticles
15	Jérôme Yon	Impact of the OC/EC ratio of soot particles generated by miniCAST and coating of these soot particles by an organic material on LII measurements
16	Jinfeng Yuan	Comparison of continuous wave and pulsed LII measurements of black carbon mass at atmospherically-relevant concentration levels

Discussion Sessions

These serve to discuss the latest developments and “burning issues” in various aspects of LII, especially those beyond the status described in literature. The material includes recent approaches or (also unexpected) results and unpublished material.

1. Determination of key parameters for LII

Organizers: Per-Erik Bengtsson (per-erik.bengtsson@forbrf.lth.se)
Eric Therssen (Eric.Therssen@univ-lille1.fr)

- Key parameters for LII: Optical properties (absorption and scattering properties); E(m), F(m), m=n-ik / Thermal accommodation coefficient / Density / Heat capacity / Particle size / Sublimation parameters / Etc.
- Change of parameters with: Maturity / Temperature / Pressure / Flame type, sources / Morphology, fractal parameters
- Methods for measuring key parameters (limitations, uncertainties)

2. Supplementary and combined techniques

Organizers: Emre Cenker (emrecenker@gmail.com)
Jérôme Yon (jyon@insa-rouen.fr)

Combined techniques aim in achieving a most complete characterization of soot particles as possible. The aggregate / primary particle size distributions, number concentration, optical/radiative properties, bulk density, composition / chemical signature, crystallinity, fine morphology (overlapping, fractal dimension, prefactor), rate of oxidation, surface growth and maturity are of interest. The discussion will deal with the coupling of optical and non-optical techniques enabling the determination of such information as well as the emerging diagnostic techniques that could be used for soot particles.

3. Pulsed and CW LII: modeling, evaluation and unresolved questions

Organizers: Kyle Daun (kjdaun@mme.uwaterloo.ca)
Raphael Mansmann (raphael.mansmann@uni-due.de)
Joel Corbin (joel.corbin@nrc-cnrc.gc.ca)

- Signal processing and analysis techniques/best practices (scaling, Q-noise removal, calibration techniques, validation of data)
- Advancements in pulsed LII modelling techniques (inclusion of other heat transfer modes, spectroscopic models, non-incandescent emission, model comparison and evaluation, etc.)
- CW-LII model development (value in using models, potential of inferring internal mixing through scattering)?
- Unexplained data features (anomalous cooling, discrepancies in LII channels, etc., abnormalities in CW-LII signals)

4. Non-soot LII

Organizer: Christof Schulz (christof.schulz@uni-due.de)

- Fundamental experiments and simulation on non-soot particles: optical properties, accommodation coefficients, evaporation properties, fluence dependence, transition to plasma emission
- Application of LII to non-soot particles
- LII combined with other laser-induced emission signals or scattering, etc.

5. Emitted and ambient aerosols

Organizers: Greg Smallwood (greg.smallwood@nrc-cnrc.gc.ca)
Francesca Migliorini (migliorini@ieni.cnr.it)

This session focuses on the experimental/numerical/theoretical activities, issues and best practice of LII applied to emitted and ambient Black Carbon.

Topics include:

- Sources and applications (soot generators, aviation, marine, and road transportation, ambient, etc.)
- Influence of BC properties on LII
  - Composition
  - Maturity
  - Optical properties (E(m), dispersion coefficient
  - Coating/mixing state
- Calibration methods/materials
- Interferences to BC measurements
- Influence of measurement methods on BC properties
- Measurement protocol
- Optimum analysis procedures (for volume, mass, primary particle diameter, and/or surface area)

Abstracts

Determination of optical properties of non-soot nanoparticles generated by microwave plasma via line-of-sight attenuation (LOSA)

M. Asif[1]**,** J. Menser[1], K. Daun[2], T. Dreier[1], C. Schulz[1]

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[2]*Department of Mechanical and Mechatronics Engineering, University of Waterloo, Canada*
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Lately, LII technique has been successfully extended to noon-soot nanoparticles [1-4]. For this purpose, the optical properties of these particles should be known to build the spectroscopic model for LII application.

In this work, a line-of-sight-attenuation (LOSA) technique has been implemented to determine the optical properties of non-soot (Ge, Si) nanoparticles. Nanoparticles are generated via gas phase synthesis by microwave plasma treatment at low pressure (100 mbar) [1]. Gases ($Ar/H_2/SiH_4$ or GeH_4) are injected with 0.03/2/0.2 slm through a nozzle into the MW plasma region of the reactor. Additionally, swirling gases (Ar/H_2) with 6.6/0.5 slm stabilize the generated particle stream.

The in-situ optical properties of the generated molten nanoparticles measured by LOSA are compared with ones obtained from literature (ellipsometry) [5,6] and calculated by Mie theory. This work also includes suggestions on solving of some current LOSA measurement issues.

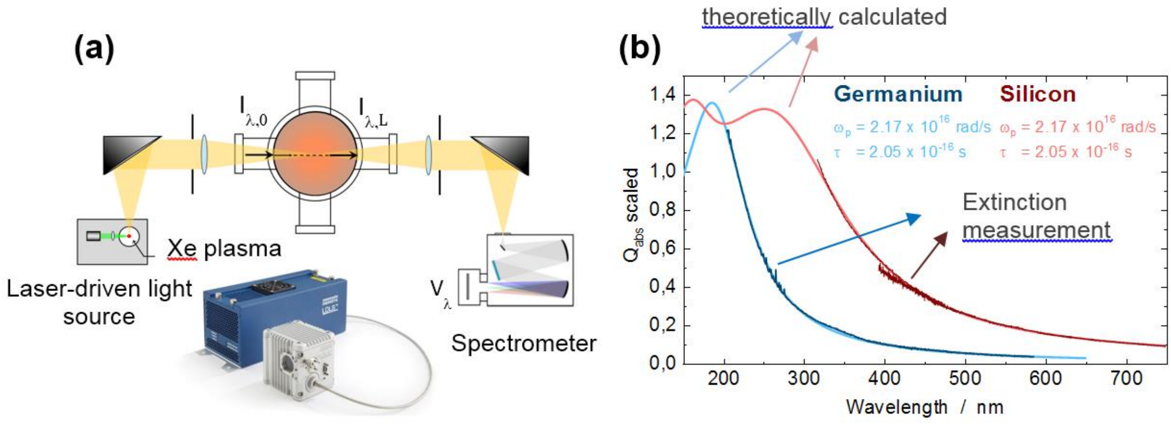

Fig. 1. **(a)** A schematic representation of the experimental setup; **(b)** Absorption spectra for Si and Ge nanoparticles measured by LOSA and theoretically calculated from Drude theory.

References

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[2] T.A. Sipkens, et al., *Appl. Phys. B*. **116**, 623-636 (2016)
[3] F. Cignoli, et al. *Appl. Phys. B*. **96**, 593-599 (2009)
[4] t. Lehre. Et al. *Proceedings of the Combustion Institute.* **30**, 2585-2593 (2005)
[5] G.E. Jellison, et al., *App. Phys. Lett*. **51**, 352 (1987)
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Investigation of soot formation in 2D by combined multi-angle light scattering and laser-induced incandescence employing a shielding approach

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Detailed investigations of the morphology of soot aggregates are crucial for a profound understanding of soot formation in combustion systems. Elastic Light Scattering (ELS) and Laser-Induced Incandescence (LII) are well-established techniques for the determination of central soot aggregate parameters in terms of the radius of gyration, the fractal dimension and the primary particle size.

The radius of gyration and the fractal dimension are determined from the angular distribution of scattered light [1]; the primary particle size is derived from the decay-time of the thermal radiation signal that occurs for cooling particles after heating them up with a short laser-pulse [2]. For a sound evaluation of experimental LII-signals, an accurate model describing heat and mass transfer between laser-heated soot particles and ambient gas is required. Depending on the aggregate size and morphology, primary particles located in the inner region of the aggregate are shielded from approaching molecules of the ambient gas resulting in a diminished heat transfer. Consequently, the radiation decay-time is increased and the primary particle size is overestimated.

We investigated the influence of the aggregate structure on the determination of primary particle size with a combination of 2D-Multi-Angle Light Scattering (2D-MALS) and 2D-LII on an extended sooting laminar premixed flame of type McKenna. Images of elastically scattered laser-light (cw-laser @532 nm) were taken in the angular range of 20° to 155° and corrected for optical distortion and extinction. Local effective values of the radius of gyration were then determined for each radial position and height above burner surface. Incandescence signals were induced with a pulsed Nd:YAG-laser (532 nm) and recorded with an intensified CCD-camera. The ratios of the signals at two different times during the cooling of the soot particle were calculated. Local effective values of the primary particle size were derived with a shielding model [3,4] considering signal ratios and local information of fluence, ambient temperature and aggregate sizes derived from 2D-MALS. The data is compared with results from a model using a constant effective thermal accommodation coefficient.

References

[1] C. Sorensen, *Aerosol Sci. Technol.* **35**, 648-687 (2001)
[2] H.A. Michelsen, C. Schulz, G.J.Smallwood, S. Will, *Prog. Energ. Combust.* **51**, 2-48 (2015)
[3] A.V. Filippov, *J. Colloid Interf. Sci.* **229**, 261-273 (2000)
[4] F. Liu, *Appl. Phys. B* **83**, 383-395 (2006)

SP2-XR: The Next Generation of Single Particle Black Carbon Instruments

A. Attwood[1]**,** H. Schulz[2], G. Granger[1], M. Zanatta[2], A.B. Herber[2], D. Baker[1] and R. Gerdes[2]

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Accurate measurements of particle concentration, composition and temporal and spatial variability are essential for estimating their radiative effects in the atmosphere. Here, we present a new single particle instrument for the measurement of black carbon using laser-induced incandescence. These particles are ubiquitous in the atmosphere and can directly affect the Earth's radiative balance by absorbing and scattering solar radiation.

Black carbon aerosol is emitted by combustion processes and plays a major role in climate forcing by absorbing solar radiation and re-radiating the energy as heat. Single particle measurements of black carbon have been developed previously but were not ideal for use in long term monitoring due to their large size and complexity of the data analysis. We have developed a new instrument, the SP2-XR, which uses the same physical principles as existing technologies but with a smaller footprint, easier data analysis and extended size range.

The SP2-XR discriminates between scattering and absorbing particles based on their interaction with a high powered Nd:YAG laser beam (Figure 1a). The signal received by the incandescence and scattering detectors can be used to calculate black carbon particle mass as well as size and number concentration of non-absorbing particles.

Previous techniques were also limited by the smallest black carbon particle size that was detectable (70 nm). Advances in the optical design and electronics allows for detection of particles down to 50 nm (Figure 1b).

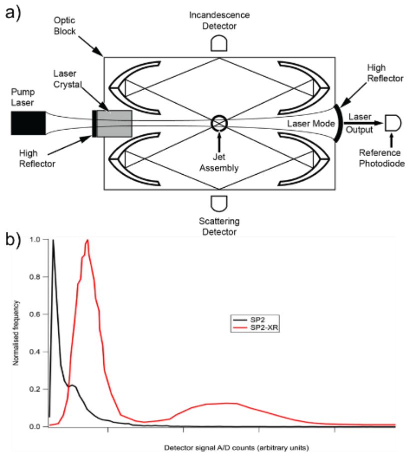

Fig. 1. a) Optical layout of the SP2-XR. b) Comparison of peak height histograms of the SP2 and SP2-XR for 56 nm Aquadag particles.

Can soot primary particle size distributions be determined using laser-induced incandescence?

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The size and morphology of nanoaggregates, both in terms of primary particle diameter, number of primary particles, and fractal structure, significantly influence their physical and chemical properties. As an example, carbon blacks, widely used as a black pigment, show a better dispersity and higher gloss for small and narrow primary particle size distributions. While aggregate parameters can be determined through light scattering [1], laser-induced incandescence (LII) is the technique of choice for *in situ* determination of primary particle size [2].

LII particle sizing relies on accurate cooling models, especially for heat conduction, to interpret the incandescence signal decay. Over the years, many concepts to determine the primary particle size distribution (PPSD) with LII have been presented, but most of these exclude the influence of the aggregate morphology on the cooling of the particles, or use an effective reduced thermal accommodation coefficient. This approach is problematic as the aggregate structure has a great impact on the cooling process by shielding the particles in the center of the aggregates [3]. To consider the shielding effect correctly, detailed knowledge about the aggregate morphology is required. Therefore, it remains questionable if the determination of the PPSD is possible when measuring polydisperse aggregate samples.

We employ Bayesian inference to answer this question. By adding prior knowledge about certain aggregate parameters – derived from different measurement techniques, e.g., wide-angle light scattering or TEM – we can evaluate probability density functions for parameters of the PPSD. A hot ambient gas case (1600 K) and a cold aerosol case (300 K) are analyzed. The results reveal the influence of uncertainties in model parameters in the LII model in the framework of the uncertainty analysis.

References

[1] C. Sorensen, *Aerosol Sci. Technol.* **35**, 648-687 (2001)
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A new approach for in situ soot size distribution measurement based on spectrally resolved light scattering.

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Air traffic increase and growing awareness of its climatic impact result in stringent regulation standards to reduce not only gaseous pollutants but also soot emission. In consequence, aircraft motorists have to develop a variety of innovative combustion systems aiming to reduce fuel consumption and soot emissions. To do this, a detailed understanding of the combustion processes and the ability to numerically simulate the combustion behaviour is mandatory and rely on accurate experimental data. A scientific cooperation between numerous academic research laboratories including the CORIA laboratory and industrial research institutes was recently established to answer those issues for the aeronautic sector (H2020 European program SOPRANO). One of the objectives is to provide accurate data on experimental pilot flames to provide a better understanding about the highly complex soot formation process in high-pressure and high-temperature operating conditions. The current study falls within this project by developing an innovative optical diagnostic able to perform in-situ measurements of soot size distribution and possibly of their optical properties. Usually, the optical diagnostics based on soot static elastic light scattering can be used but they provide a partial determination of the particle size distribution [1] or require a detection system composed of detectors placed at a minimum of three angles [2]. Because the high-pressure optical combustion chambers generally offer limited optical accesses, there is therefore also a need for a two-angle optical technique to be developed. In the current study, the measurement strategy consists in filling the gap caused by the lack of available detection angles by recording the spectral response of the static light scattering.

We present a proof of concept of the spectral light scattering analysis on well-known soot particles produced by a commercial soot generator (CAST). The Rayleigh-Debye-Gans theory for Fractal Aggregates (RDG-FA [2]) is applied to process the light scattering spectra and therefore to obtain information on the mode and standard deviation of the studied polydisperse size distribution. The data processing principle is based on the ratio of the light scattered signals at two angles in order to remove the dependency of the measurement on parameters such as the fractal prefactor, particles primary diameter, aggregates number density and soot optical properties. The results are then compared to classical SMPS measurements.

Once the size distribution is determined, the light scattering spectra are analysed again to obtain the spectral evolution of the soot optical properties (F(m) in the RDGA-FA theory, with m being the complex optical index). This is of interest since it has been shown that the spectral dependency of F(m) is related to the soot constituents and structure [4].

References

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LII particle-size imaging with an ultra-high-speed CMOS camera

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Quantitative particle-size information can be obtained from a best-fit comparison of the temporal laser-induced incandescence (LII) signal decay and simulations based on the particles' energy and mass balance equations. Similarly, spatially resolved particle-size information can be deduced by comparing the local ratio of temporally-distinct LII signals from two or more sequential images against the simulations. Historically, intensified CCD cameras have been the workhorse for LII imaging due to their ultra-short gating capabilities and sensitivity to light. With their slow read-out times, however, a single CCD detector cannot provide consecutive images within the typical LII signal lifetime. Consequently, acquiring temporally resolved LII images required either multiple cameras or a time-gate sweeping strategy relative to the laser pulses, which is limited to steady or repetitively pulsed flame conditions.

Time-resolved 2D LII signals were acquired with an ultra-high-speed, non-intensified, CMOS camera at 10 million frames per second (fps) and 100 ns gate width. The proof of concept measurements used a co-annular non-premixed laminar ethylene/air flame (Santoro burner). Particles were heated with a 1064-nm (injection seeded) laser sheet operated at fluences from 0.03 to 0.15 J/cm^2. The first 10 to 20 images following laser excitation show LII signal that is clearly above the background signal and sufficiently strong to compare to the modeled LII decay (see Fig. 1). The LII signal decay rate at various flame heights along the flame edge were extracted and compared to previously published decay rates acquired with a photo-multiplier tube (PMT) from the same nominal flame. Signals from the ultra-high-speed camera are in good agreement with the previous PMT measurements. The signal decay profiles at all laser-heated locations were used as inputs to the LII model for quantitative particle sizing. Other relevant model inputs such as flame temperature, aggregate size and the soot absorption function were imported from the literature. Evaluated particle sizes show good agreement with those reported in the literature at various flame heights. Compared to imaging with multiple sensors or time-gate sweeping strategies, this new ultra-high-speed LII imaging reduces uncertainties due to image mapping, flame perturbations and laser shot-to-shot variations. In the second phase, experiments will be performed in a turbulent flame with the objective of providing instantaneous spatial distributions of soot particle sizes at various Reynolds numbers.

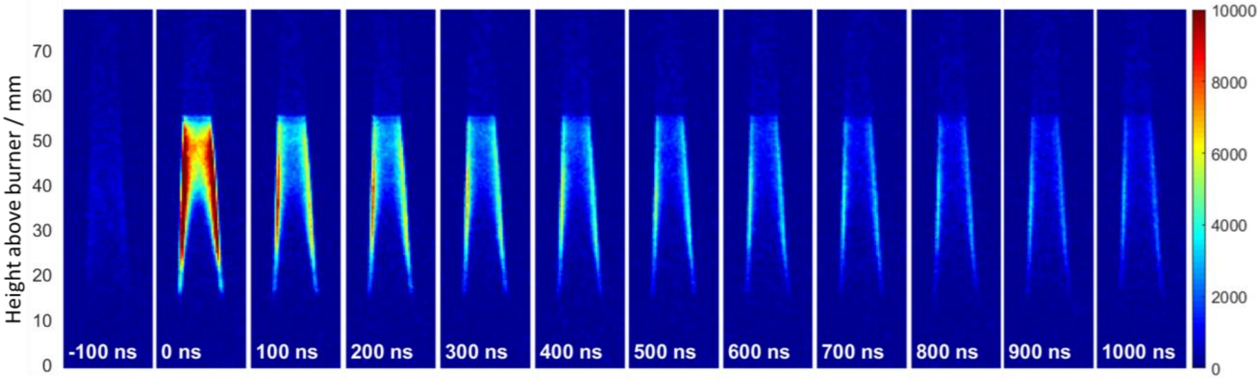

Fig. 1 Flame images before and after heating with a laser pulse at 0.08 J/cm^2. Time stamps show the delay with respect to the laser pulse.

Effects of soot volume fraction on bath-gas heating and particle sizing during LII

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During laser-induced incandescence (LII), energy absorbed by particles transfers to the surroundings leading to an increase in temperature of the bath gas. In a quantitative soot particle sizing approach based on LII modeling, the rise in the bath-gas temperature can significantly influence results. Two-color pyrometry imaging is performed to measure this temperature increase at five different soot load conditions in a non-premixed laminar nitrogen diluted ethylene flame. Soot particles are heated volumetrically at 11.3 mm height above the burner with a 1064 nm laser at two different fluences with a 3-ns long quasi-tophat pulse. Images were acquired at 5 and 10 µs after the laser pulse. The soot load in the flame is altered by changing the ambient pressure from 2 to 10 bar. The fuel and nitrogen flow rates are adjusted to keep the visible flame height at 18±1 mm at all conditions. Line-of-sight averaged soot volume fraction (SVF) is measured via light extinction method (LEM) with a CW laser at 632 nm. The lowest and highest SVF are 2.6 ppm and 31.7 ppm, respectively for varying conditions at the flame height where pyrometry measurements are performed. Measurements show that the bath-gas temperature after the LII increases drastically with increasing SVF due to the increased total amount of transferred energy. To simulate this change of bath-gas temperature, a theoretical sub-model based on tracking of energy transferred via conduction and adiabatic mixing of evaporated soot is implemented into an existing LII model. The simulations are run for different SVF conditions. The measured and simulated residual temperatures are in good agreement. The quantitative effects of bath-gas heating on particle sizing for various soot load scenarios are calculated with the LII model.

Turbulent flame LII particle sizing via ultra-high-speed imaging

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Soot particle size estimates can be obtained by measuring the decay time of laser induced incandescence (LII) signals in flames. Current state-of-the-art methods for making these measurements in 2D require intensified CCD cameras with slow read-out times. Thus, temporally resolved LII can only be acquired with multiple cameras or time-gate sweeping with multiple laser pulses. Therefore, existing methods are best suited for steady flame conditions. However, with the use of new high-sensitivity ultra-high-speed cameras, single-laser-shot 2D soot particle sizing is now possible, opening the door to measurements in turbulent flames and other unsteady combustion regimes. In this work, time-resolved LII signals were acquired using an ultra-high-speed non-intensified CMOS camera (Shimadzu HPV-X2, 10 million frames per second, 100 ns exposures, 256 frames). Particles were heated using a ~10 nanosecond, 1064 nm injection seeded laser. First, the camera was fully characterized and validation experiments were conducted using a Santoro burner (co-annular non-premixed laminar ethylene–air flame), showing that measured decay rates and predicted particle sizes matched literature values. Then, measurements in a turbulent ethylene jet flame generated using a Shaddix burner (ethylene–air pilot, D = 3.2 mm) were performed at varying heights, laser fluences, and Reynolds numbers. Preliminary results of the first few frames captured after the laser pulse, shown in Fig. 1, indicate that the decay rates of the soot incandescence differs throughout the images, further suggesting that soot particle sizes vary throughout the turbulent flame. These non-intensified images show that there is sufficient camera sensitivity to capture soot incandescence at laser fluences well below sublimation levels. Signal decay profiles are then measured and compared with an LII model combined with literature estimates of flame temperatures and soot absorption functions. Compared to alternative multi-camera strategies, measurements with the ultra-high-speed camera do not require registering pixels across cameras, thus reducing uncertainties and setup complexity.

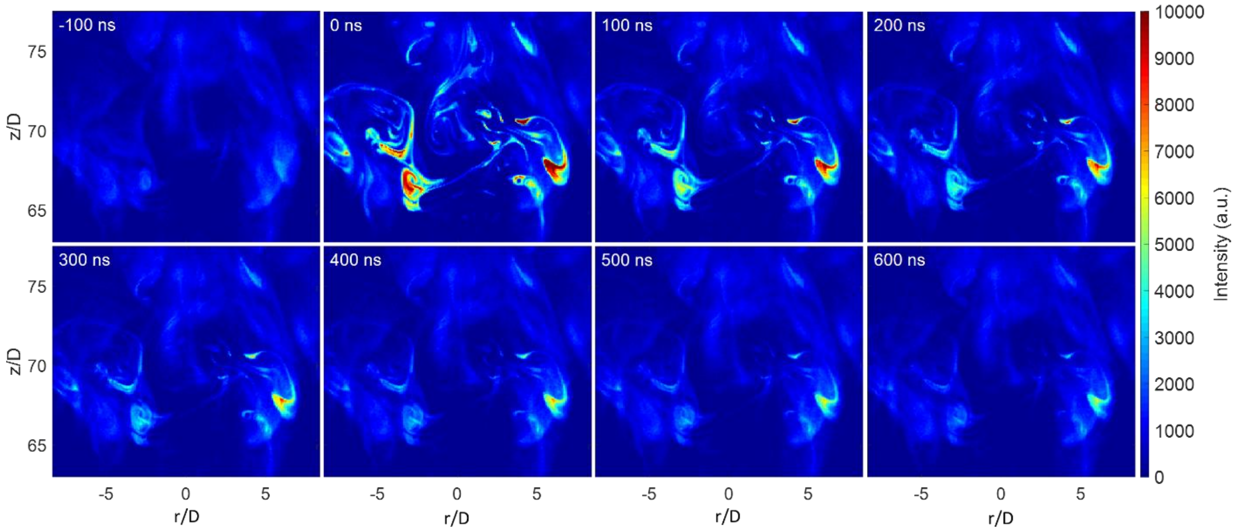

Fig. 1. Turbulent flame LII imaging before and after laser heating with a fluence of 0.15 J/cm^2 for a jet with Re = 20,000. Images are obtained through a 608–674 nm bandpass filter. The timing relative to the laser pulse is shown in each image.

Laser-induced incandescence of aircraft engine black carbon: sensitivity to laser fluence

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The calculation of BC mass concentrations from LII measurements requires knowledge of the absorption function E(m), which is well known to vary between black carbon (BC) samples [1], reflecting the fact that the molecular structure (e.g. degree of graphitization) of BC varies between sources. Related to this variation, the optimal laser fluence for use in LII has been determined as that where particles are heated to incandescence (> 3000 K) while avoiding carbon sublimation at ~4500 K. However, this ideal fluence may differ between BC sources, so that the behavior of laboratory-flame BC may differ from that of aircraft-engine BC. To directly constrain this issue, we directly examined the dependence of LII-measured BC concentrations on laser fluence for aircraft engine BC produced by a variety of fuels and engine thrust conditions.

We performed measurements as part of the ECLIF-2/ND-MAX campaign at the Ramstein Air Base, Germany. During this study, the V2527 engines of the DLR A320-232 "D-ATRA" research aircraft were operated with two different conventional fuels (Jet A1) and three fuel blends of conventional jet fuel and sustainable alternative jet fuel (SAJF) at a range of N1 fan speeds ranging from idle (25%) to maximum continuous thrust (82%). Two Artium LII-300 instruments measured BC mass concentrations in the aircraft engine exhaust. One instrument was configured as the reference, operating at a laser fluence optimized for aircraft engine BC. The laser fluence of the second instrument was varied non-linearly from full laser fluence to very-low laser fluence by varying the Q-switch delay time. The full fluence data were used as a reference to determine periods of stable BC concentration.

The sensitivity of LII-measured BC to laser fluence was quantified as the ratio of the modified signal to the reference signal, where the modified signal was the low-fluence condition (longer Q-switch delay) and the reference signal was the high-fluence condition (minimum Q-switch delay). We investigated this ratio as a function of Q-switch delay. The ratio fell below unity at Q-switch delays of about 163 µs for measurements at engine idle (25% N1 fan speed), a substantially longer delay than the full-fluence value of 143 µs. This fall-off point became systematically longer at higher thrusts, up to 82% N1 fan speed. Data for five fuels indicates that the variation between engine thrust conditions was greater than the variation between fuels. These measurements demonstrate that the response of LII to aircraft-engine BC emissions is independent of the engine thrust under full fluence conditions, supporting the use of LII in quantifying those emissions.

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Coupling of cavity-ring-down extinction and laser induced incandescence to determine soot volume fractions in a nucleation and a sooting premixed flames

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Understanding of soot formation, particularly the nucleation step leading to the formation of the nascent soot particles from gaseous precursors, is critical to develop reliable predictive soot models and to help the design of more efficient and cleaner combustion devices.

This presentation focuses on the recent advances achieved through detecting soot nanoparticles of size 2-4 nm using laser-induced incandescence(LII) [1-4]. These particles are found in the nucleation zone of sooting flames or all along the so-called nucleation flames. In this work we define a particle as soot if the particle absorbs radiation and emits blackbody-like radiation in the visible and infrared. The objective of this work is to conduct quantitative measurements of soot particles at very low concentrations. The increasingly accomplished improvement of the LII technique led recently PC2A to investigate its highest sensitivity to detect the lowest possible soot volume fraction in sooting flames, notably by gradually decreasing (from a typical sooting flame condition) the equivalence ratio of the flame at the limit of the characteristic visible yellow soot luminosity.

The decrease in the equivalence ratio was also accompanied by a gradual decrease of the LII decay time until a constant value. This decay time is related to the cooling time of the laser heated particles and thus to the size of the incandescent particles. From this strategy performed in flames with different fuels and pressures we proved the existence of flames producing nascent soot particles that do not show a measurable diameter increase along the height above the burner [1-4]. We named these flames *nucleation flames* to highlight that the soot mass increase is mainly attributed to an increase of the number of nucleation soot along the flame, but not from surface growth. From modelling the LII decay time and making some assumptions on the physical-chemical parameters of these particles based on the experimental observations, the diameter of nascent particles was estimated around 1- 3 nm, in very good agreement with ex-situ size determination by nano-scanning mobility particle sizer (SMPS) [4]. In this work, LII was calibrated to provide absolute soot volume fraction using cavity ring-down extinction (CRDE). From combined LII and CRDE measurements, and LII modelling an original methodology is proposed that allows to minimize the impact of the lack of knowledge of the spectral variation of soot absorption function E(m) with soot maturity. Finally, the absolute soot volume fraction profiles were obtained over a very large dynamic range in several atmospheric flames. These data are very useful for soot modelling validation [5-6].

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Thermographic and two-phase PIV based on LII signal from submicron black particle tracers

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We consider the application of laser induced incandescence (LII) of submicron particles two purposes: (a) as a thermographic particle for simultaneous velocity and temperature measurement, and (b) as a method for two-phase flow velocimetry. In thermographic Particle Image Velocimetry (PIV) phosphorescent particles have been used achieve simultaneous temperature and velocity imaging in gaseous flows. However, the high cost for phosphor materials and the low signal yields obtained at high temperatures limit the application of this technique. In the first part of this paper, we investigate the possibility of using two-colour (2C) measurements from the laser-induced incandescence (LII) signal using submicron black particle tracers. These particles are dispersed into a flame and further heated by a double-pulsed top-hat laser sheet. Unlike conventional LII, energies are kept below the limit of sublimation, so that the gas temperature can be indirectly measured by subtracting the calibrated temperature increase ΔT due to laser absorption from the measured particle temperature. The particle peak temperature can be measured by two-colour pyrometry using two ICCD cameras, while the laser-induced temperature increase ΔT can be determined by a calibration at a reference local gas temperature. The velocity field can be extracted from the cross-correlation of two consecutive frames using either the same intensified images (at lower resolution) or a third camera. The feasibility of the proposed technique is demonstrated by a numerical model of the particle heating and LII signal emission intensity as a function of time and spectral characteristics, parameterised by the flow temperature and particle diameter. Numerical results suggest that the combination of a wide bandpass filter in near the UV range and a narrow band pass filter in near the IR region is the best option in terms of temperature sensitivity and signal detectability for ICCD cameras. The signal ratio under such case is highly sensitive to the local gas temperature beyond 1200 K.

In a second study, we demonstrate the use of laser induced incandescence (LII) of submicron tungsten carbide (WC) particles as a method for particle image velocimetry (PIV). The technique allows a single laser to be used for separate measurements of velocity of two phases in a droplet-laden flow. Submicron tungsten carbide (WC) particles are intentionally seeded into a two-phase flow, and heated by a light sheet generated by a double-pulsed PIV laser operating at sufficiently high pulse energy. The small size and large absorption cross-section allows particles to be heated up to several thousand degrees to emit strong incandescence signals, whilst the laser-induced temperature increase in liquid droplets/large particles is negligible. The incandescence signal from WC and Mie scattering from droplets/large particles are separately captured by deploying different filters to a PIV camera. The consecutive images of the laser-induced incandescence (LII) are used to determine the velocity field of the gas-phase flow, and those of Mie scatter are used to extract the velocity of droplets/large particles. The proposed technique is demonstrated in an air jet first and compared with the result given by a normal PIV test, which shows that submicron WC particles can accurately follow the gas flow, and that the LII images can be used to perform cross-correlations. We then apply this technique on an ethanol droplet/air jet (non-reacting), demonstrating the resulting slip velocity between two phases. The proposed technique combining PIV and LII with a single laser is applicable to a much higher droplet/particle density than previously feasible.

LII and MAE measurements in a laminar diffusion flame to assess the ISF database consistency

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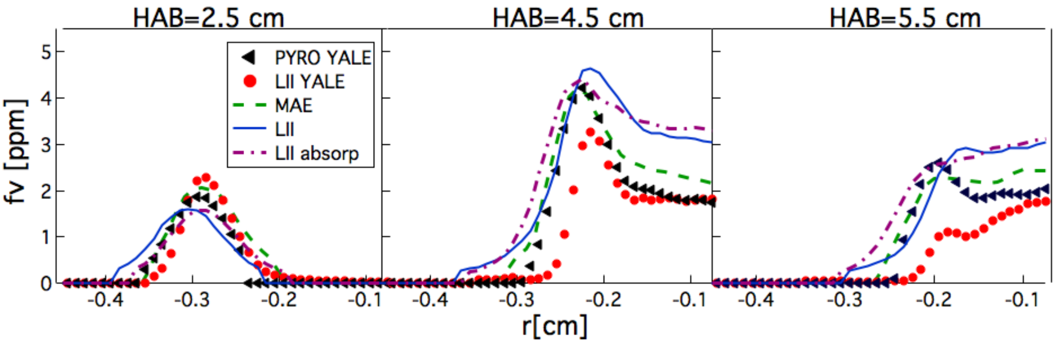
The control of soot emission raises fundamental issues and has important practical implications requiring a full understanding of soot production and oxidation processes. In this context, the International Sooting Flame workshop (ISF) aims at studying various sooting configurations both numerically and experimentally. Data are shared to improve the precision and the validity of the different experiments. The research reported in this work aims to contribute to the studies carried out on laminar sooting flames which are indeed crucial for the validation of numerical models before their extension to turbulent configurations. The objective is to quantify sources of experimental errors and to extend the existing database for the Yale laminar diffusion burner flame to enable more comprehensive comparisons among different experimental techniques and numerical simulations. For this, a combined use of MAE[1] (Modulated and Absorption Emission) and LII techniques is presented in this work. MAE is here used to calibrate the LII. In addition, a method to estimate LII self-absorption is proposed based on the MAE deconvolution technique and Time-Resolved LII measurements are performed to obtain information on the primary particle size distribution. Results are compared with already existing experimental data highlighting the high variability of the experimental data depending on the measurement techniques as well as the underlying assumptions and post-processing methods. Fig. 1. Soot volume fraction at three heights above the burner for the 80% case. Comparison of different measurement techniques for fv (LII from Yale [2], pyrometry from YALE [3], new LII[4] and MAE[4] results).



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A calibration strategy for planar laser-induced incandescence measurements at increased pressure

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Planar laser-induced incandescence (PLII) is a valuable tool to determine soot distributions in generic laboratory flames and technical combustion devices, characterized by increased pressure. Quantification of PLII is either realized via calibration with a calibration flame of known soot concentration, placed in the location of the measurement [1], or by an independent diagnostic such as extinction [2] or gravimetric sampling [3]. Gravimetric sampling or application of an additional extinction measurement at increased pressure is far from trivial if instantaneous calibration is planned, and the simple transfer of calibration information from atmospheric conditions to high pressure is expected to introduce errors. This is the result of a change of the signal decay behavior with increasing importance of conduction at higher pressures (Fig. 1), and varies with the choice of the detector gate duration.

We use an exemplary model [4] and experimental excitation and detection characteristics [5] to quantify this effect for a pressure range which appears reasonable for laser-based gas turbine studies. This involves a laser fluence of 0.4 J/cm^2 at 1064 nm, for simplicity a top hat laser beam profile (although the sheet thickness in typical imaging LII applications is rather Gaussian in shape), the measured laser temporal profile and LII filter transmissivity curve, and finally a representative gate of 60 ns. The modelled LII signal is consequently integrated for this gate period starting from the onset of detectable LII signal. Fig. 2 shows the resulting signal decrease for increasing pressure. Different quantities affecting this procedure are considered such as particle size, use of a size distribution, and respective LII fluence curves are taken into account.

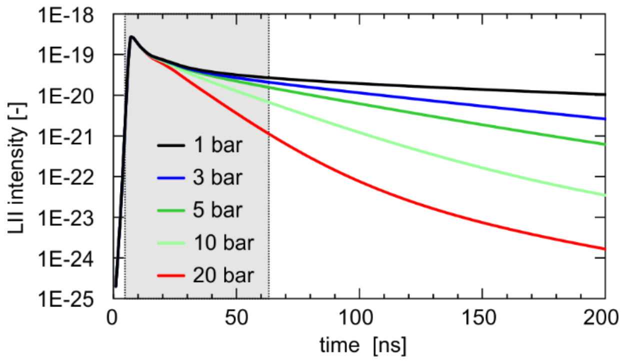

Fig. 1. Variation of signal decay time with pressure for a monodisperse particle of 30 nm, ambient gas temperature and employed fluence of 0.4 J/cm^2 at 1064 nm.

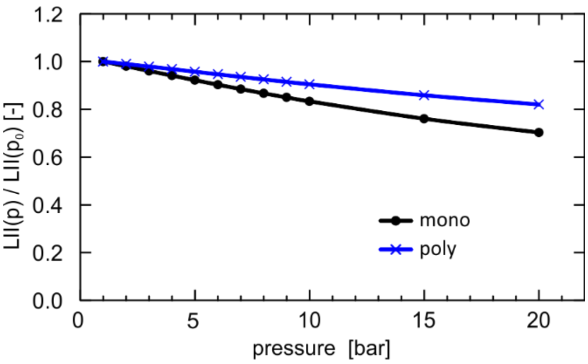

Fig. 2. Decrease of signal emitted by the same particle with pressure and prompt signal integration over 60 ns for monodisperse particle and polydisperse distribution.

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Photoacoustic Measurement of Soot in a Flat Flame with a High Rep Rate Fibre Laser

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We have previously used a high-power cw diode laser to measure soot distribution in a flat flame burner using the photoacoustic effect, and compared the results with prompt LII [1]. Here we will present photoacoustic measurements in the same burner using a commercial high-repetition rate fibre laser (SPI Lasers G4), which has previously been used for LII measurements in a helicopter engine exhaust [2]. The laser is better suited for photoacoustic measurement using lock-in detection as it produces pulses approximately 200 ns long at 10's of kHz repetition rates, effectively producing a low duty cycle on/off modulation at frequencies that fall within the operating range of commercially available microphones. This is an advantage compared to the diode laser used previously, which produced a small modulation depth despite having a relatively high average power. In fluence curves, the photoacoustic signal shows the 'plateau' behaviour typical of LII in the particle sublimation regime, though the level of photoacoustic signal at which the plateau occurs increases with repetition rate. At high repetition rates particles undergo a number of interactions with the laser energy as they move through the measurement volume, producing a photoacoustic signal at higher harmonics of the repetition frequency (Fig. 1). Future experiments (both LII and photoacoustic) using longer wavelength and continuous wave fibre lasers are planned.

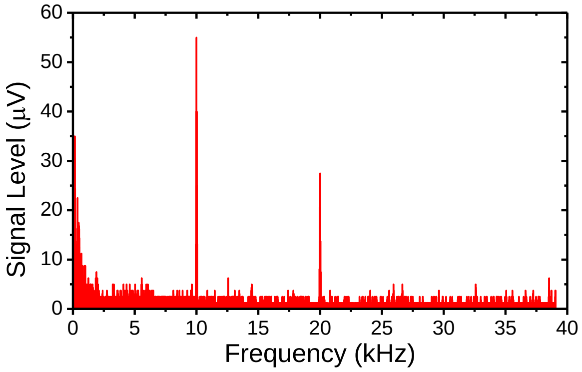

Fig. 1 - FFT of raw (uncorrected) microphone output for a 10 kHz pulse repetition-rate (laser power setting of 80%)

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Multi-diagnostic imaging of evaporating fuel wall-films in combustion as a source of PAH and soot

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In direct-injection gasoline engines, evaporating fuel wall-films and the resulting inhomogeneities of the air-fuel mixture near those films make the formation of PAH and soot in subsequent combustion likely. Different optical techniques are needed to visualize the links of this process chain, such as the spray, film formation, evaporation, combustion, and soot formation. In our model experiment, shown in Fig. 1, a mixture of isooctane (surrogate fuel) and toluene (fluorescent tracer) is injected by a multi-hole injector into an optically accessible flow channel. Air flows continuously through the channel at room pressure. Combustion is initiated by a spark plug within the fuel/air-mixture cloud. Some of the liquid fuel impinges on the quartz-glass wall on the opposite side and forms wall films. The turbulent flame front propagates along the chamber and ignites pool fires above the wall films, leading to locally sooting combustion. Laser-induced fluorescence (LIF) of the toluene using 266 nm excitation images the fuel-film thickness and visualizes the fuel vapor above the liquid films. Laser-induced incandescence (LII) using 1064 nm excitation visualizes soot. As a complementary visualization of soot, the natural flame luminosity, mainly from soot incandescence, is captured with a high-speed camera. Schlieren imaging combines the visualization of the evaporating liquid and the sooting flame. The LIF images show that indeed the fuel wall-films remain on the surface long after the flame front has passed, leading to subsequent soot formation. In a next step, we will simultaneously visualize the formation of soot precursors (PAH) and soot with LIF and LII, respectively.

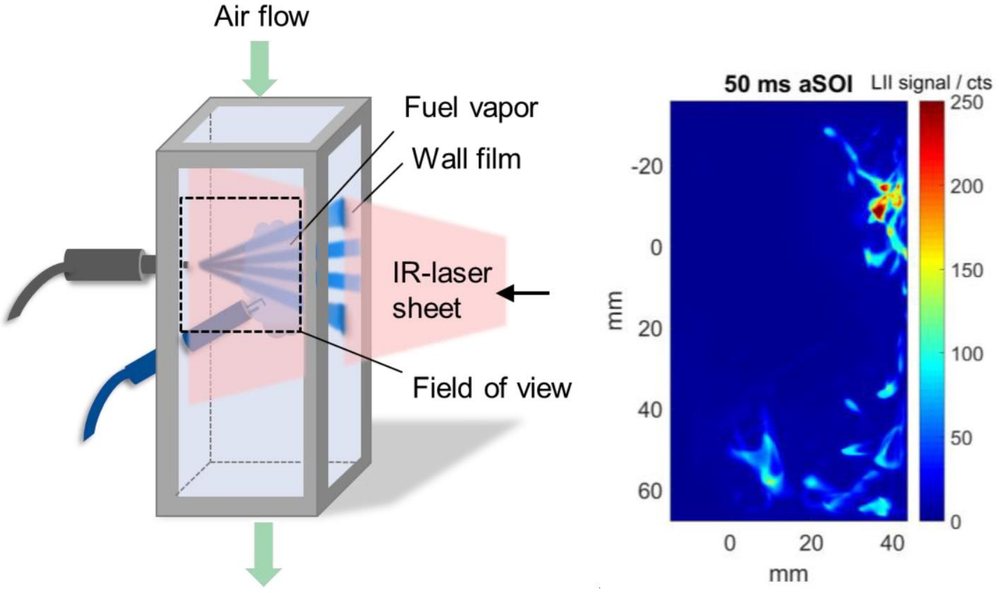

Fig. 1. Experiment (left), LII of soot from bottom and top wall-film (right).

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Raman spectroscopy on soot produced from a mini-cast soot generator: impact on structure from heating in air and nitrogen up to 900°C

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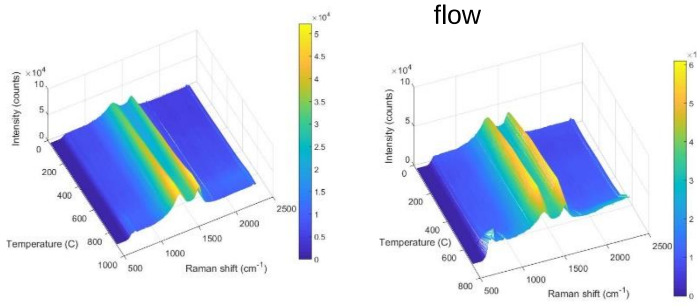
Soot is formed from incomplete combustion and can, when freshly emitted to the atmosphere, show a variety in its characteristic properties: size, morphology, internal structure, optical properties. The characteristic properties of soot depend on various parameters in the combustion process such as the fuel and type of combustion process as well as on the time and temperature history. In this work, a study was made to improve understanding of soot nano-structures and soot oxidation processes. A mini-CAST soot generator was used to produce five soot types named OP1, OP3, OP5, OP6, and OP7 spanning from larger particles of mature character to smaller soot particles of immature character[1]. Then the sampled soot was heated in air and an inert N_2 atmosphere up to 900 ^{0}C by a LINKAM heating stage. Via Raman spectroscopy, optical and structural studies were performed on the soot samples during these heat treatments to study the effects of the air interaction on soot structures via oxidation processes, and how the internal bonding structure of soot is affected in order to gain knowledge about maturation in soot formation processes.

Figure 1 demonstrates the spectral evolution of the representatives of two soot types heated in N_2 and in air. In the 700 to 2400 cm^{-1} spectral range, the typical D and G Raman bands at about 1350 and 1600 cm^{-1}, respectively superimpose on fluorescence. Several interesting results were also obtained regarding evaporation of the organic fraction, re-structuring of soot with increasing temperature, and formation of fluorescing species during heating processes.

a) OP1 heated in N_2 flow b) OP1 heated in air flow



c) OP7 heated in N_2 flow d) OP7 heated in air flow

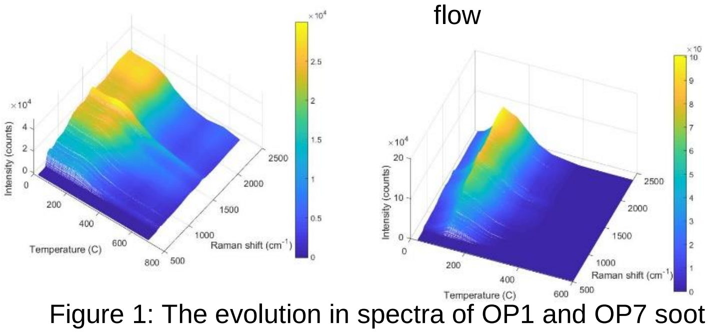


Figure 1: The evolution in spectra of OP1 and OP7 soot samples heated in N_2 and in the air environments

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Impact of the OC/EC ratio of soot particles generated by miniCAST and coating of these soot particles by an organic material on LII measurements

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There is currently a strong interest in developing optical diagnostics for black/brown carbon emissions into the atmosphere. Indeed, optical techniques enable on-line characterization that largely avoids the problems induced by physical sampling, diluting and other intrusive processes. Optical techniques, such as laser-induced incandescence (LII), light scattering, and light extinction, have long been developed and employed for the characterization of in-flame soot particles. It therefore appears promising to apply such techniques to characterize particulate matter (PM) during their aging process in the atmosphere. Regardless of the source of PM emissions (aero-engines/cars/industrial combustion systems), once emitted into the atmosphere, soot particles interact physically and chemically with the ambient volatile organic compounds and/or other atmospheric aerosols that affect their morphological and optical properties. For example, it was shown that the organic coating can cause a strong restructuration of fractal aggregate soot particles [1] and also an enhancement of its light absorption and scattering properties [2]. In consequence, some studies have to be performed in order to ensure the reliability of such techniques for their applications for atmospheric concerns.

The present work concerns the application of the LII technique to both freshly generated soot particles by a miniCAST generator and coated soot particles. The miniCAST enables the generation of particles whose morphological and optical properties have been already characterized for different operating conditions. Firstly, three operating conditions of the miniCAST were investigated because of their different OC/TC ratios. Secondly, in order to simulate aged particles that have acquired coating of organic compounds in the atmosphere, a coating of oleic acid was added to mature soot produced by the miniCAST generator. The coating thickness was varied by controlling the temperature of the oleic acid in the coating chamber. The application of the LII technique to coated particles has already been conducted showing to a certain extent a shift of the normalized fluence curves [3].

In the present study, the time-resolved two-color detection (532 and 632 nm collected by two photomultiplier tubes) and spectrally resolved emissions (over 250 ns, covering the 500-700 nm domain collected by a spectrometer coupled to an intensified camera) were investigated. In both cases the fundamental of a Nd:YAG laser, shaped into a top-hat profile, interacts with freshly generated particles. An integrating sphere was used to calibrate the devices and to provide the absolute radiance.

The maximum radiance and the associated decay time will be reported as a function of the laser fluence for different sources of soot particles, coating thicknesses, and fluences. It will be demonstrated that the core soot OC/TC ratio has a strong influence on the measured maximum radiance and decay time. First attempts for the modelling of the 2C-LII temporal and spectral signals by a simple LII model will be presented.

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Effect of detection wavelengths on soot volume fraction measurements using auto-compensating (two-color) LII

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Laser-induced incandescence (LII) has evolved as a powerful diagnostic for soot volume fraction (SVF) and soot particle size measurements both inside flames and at engine exhaust. Due to various issues associated with the conventional LII technique for quantitative measurements of SVF, such as changes in the local gas temperature, soot particle size, and pressure, the auto-compensating LII (AC-LII) or two-color LII (2C-LII) has become the preferred approach to conduct quantitative LII measurements of SVF, since it can largely compensate the changes mentioned above by inferring the effective soot temperature in the detection volume.

Although the effect of the two detection wavelengths on the sensitivity and relative error of AC-LII has been discussed by Liu et al. [1], there has been no attention paid to the effect of detection wavelengths on the inferred effective soot temperature based on the principle of two-color pyrometry and consequently SVF under conditions of non-uniform soot particle temperatures in the detection volume. The temperatures of soot particles in the LII detection volume are in general non-uniform in practical applications for various reasons, such as the spatial laser energy distribution across the laser beam cannot be made perfectly top-hat, primary soot particles are always polydispersed, and the size-dependent effect of organic coating on soot particles. Consequently, the effective soot temperature inferred from AC-LII is dependent on the two detection wavelengths, which in turn affects the inferred SVF.

In this study, the effect of detection wavelengths on the effective soot temperature and inferred SVF under conditions of both in-flame and quenched flame using a typical AC-LII set-up was numerically investigated. Soot was excited thermally with a pulsed Nd:YAG laser operated at 1064 nm. LII signals were detected at three wavelengths centered at λ_1 = 445 nm, λ_2 = 692 nm, and λ_3 = 797 nm. The laser fluence histogram without the use of a diffraction optical element has been described in [2]. The LII model used in this study was essentially the same as that employed in [2].

The results show that the effect of detection wavelength is relatively small in in-flame measurement, but becomes fairly significant in the quenched flame measurement. The effective soot temperature is more biased toward the peak temperature using the signals detected at λ_1 and λ_3, leading to a more significant underestimate of the soot volume fraction. For accurate measurements of soot volume fraction using AC-LII, two longer detection wavelengths, such as λ_2 and λ_3, are recommended to minimize the effect of non-uniform soot temperatures in the detection volume.

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LIISim: A modular signal processing toolbox for laser-induced incandescence measurements

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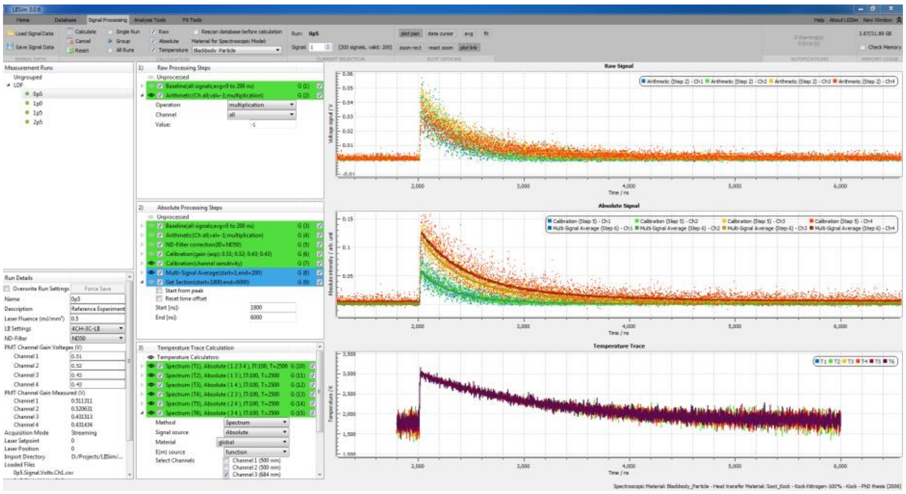
Evaluation of measurement data for laser-induced incandescence (LII) is a complex process that involves many processing steps starting with import of data in various formats from e.g., an oscilloscope, signal processing for converting the raw signals to calibrated signals, application of models for spectroscopy/heat transfer and finally visualization, comparison, and extraction of data.

We developed a software tool for the LII community that helps to evaluate, exchange, and compare measurement data among research groups and facilitate the application of this technique by providing powerful tools for signal processing, data analysis, and visualization of experimental results. A common file format for experimental data and settings simplifies inter-laboratory comparisons. It can be further used to establish a public measurement database for standardized flames or other soot/synthetic nanoparticle sources. The open-source concept and public access to the software development should encourage other scientists to validate and further improve the implemented algorithms and thus contribute to the project.



☑ Raw
☑ Absolute
☑ Temperature
○ Rescan database before calculation
Material for Spectroscopic Model: Blackbody Particle
Run: 0p5
Signal: 1 (200 signals, valid: 200)
3.87/31.89 GB

Measurement Runs
Ungrouped
LDF
0p5
1p0
1p5
2p0

1) Raw Processing Steps
Unprocessed
☑ Baseline(all signals;avg=0 to 200 ns) G (1) ☑
☑ Arithmetic(Ch all;val=-1;multiplication) G (2) ☑
Operation: multiplication
Channel: all
Value: -1

2) Absolute Processing Steps
Unprocessed
☑ Baseline(all signals;avg=0 to 200 ns) G (3) ☑
☑ Arithmetic(Ch all;val=-1;multiplication) G (4) ☑
☑ ND-Filter correction(ID=ND50) G (5) ☑
☑ Calibration(gain (exp): 0.51; 0.52; 0.43; 0.43) G (6) ☑
☑ Calibration(channel sensitivity) G (7) ☑
☑ Multi-Signal Average(start=1;end=200) G (8) ☑
☑ Get Section(start=1800;end=6000) G (9) ☑
☐ Start from peak
☐ Reset time offset
Start [ns]: 1800
End [ns]: 6000

3) Temperature Trace Calculation
Temperature Calculators
☑ Spectrum (T1), Absolute (1 2 3 4), IT:100, T=2500 G (10) ☑
☑ Spectrum (T2), Absolute (1 3), IT:100, T=2500 G (11) ☑
☑ Spectrum (T3), Absolute (1 4), IT:100, T=2500 G (12) ☑
☑ Spectrum (T4), Absolute (2 3), IT:100, T=2500 G (13) ☑
☑ Spectrum (T5), Absolute (2 4), IT:100, T=2500 G (14) ☑
☑ Spectrum (T6), Absolute (3 4), IT:100, T=2500 G (15) ☑
Method: Spectrum
Signal source: Absolute
Material: global
E(m) source: function
Select Channels:
☐ Channel 1 (500 nm)
☐ Channel 2 (500 nm)
☑ Channel 3 (684 nm)

Run Details
☐ Overwrite Run Settings
Name: 0p5
Description: Reference Experiment
Laser Fluence (mJ/mm²): 0.5
LII Settings: 4CH-3C-LII
ND-Filter: ND50
PMT Channel Gain Voltages (V)
Channel 1: 0.51
Channel 2: 0.52
Channel 3: 0.43
Channel 4: 0.43
PMT Channel Gain Measured (V)
Channel 1: 0.511311
Channel 2: 0.520631
Channel 3: 0.431513
Channel 4: 0.431434
Acquisition Mode: Streaming
Laser Setpoint: 0
Laser Position: 0
Import Directory: D:/Projects/LIISim/...
Loaded Files
0p5.Signal.Volts.Ch1.csv

Spectroscopic Material: Blackbody_Particle - Heat transfer Material: Soot_Kock - Kock-Nitrogen-100% - Kock - PhD thesis (2006)

Fig. 1. Graphical user interface of signal processing module

The public release of LIISim 3.0.6 (including user guide and example data) can be downloaded from www.liisim.com. We encourage all interested scientists to test and discuss the software on the workshop and give us feedback for future developments.

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Transition from laser-induced incandescence (LII) to laser-induced breakdown spectroscopy (LIBS) on elemental nanoparticles

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Interpretation of laser-induced incandescence data requires a heat transfer model. Due to large uncertainties in thermophysical properties, researchers avoid temperature regimes where evaporation is dominant. Most heat transfer models also exclude direct simulation of laser heating, and instead focus on the return of the nanoparticles to thermal equilibrium with the bath gas starting from a pyrometrically-inferred peak temperature. As consequence, most LII studies are limited to low laser fluences, with an emphasis on inferring aerosol properties like particle diameter and volume fraction from the nanoparticle cooling rate.

In contrast, this work focusses on laser heating in a Ge nanoparticle aerosol in the high-evaporation regime, where additional signal components from emitted atoms, electrons and ions provide rich information about the peak temperature, evaporation rate, and nanoparticle composition, beyond what is possible using only nanoparticle incandescence. Assuming thermal equilibrium, signal intensities can be related to the particle temperature and the atom concentration that is linked to the evaporation rate, cf. Fig. 1 [1].

This information can be used to infer fundamental thermodynamic properties of the nanoparticle material, including the Antoine coefficients relevant for the saturation vapor pressure, with uncertainties equivalent to, or lower than, comparable values reported in literature. Generally, this work highlights the application of a combined LII and LIBS measurement for promoting the knowledge of thermophysical properties of nanoparticles to more complex systems and higher temperatures.

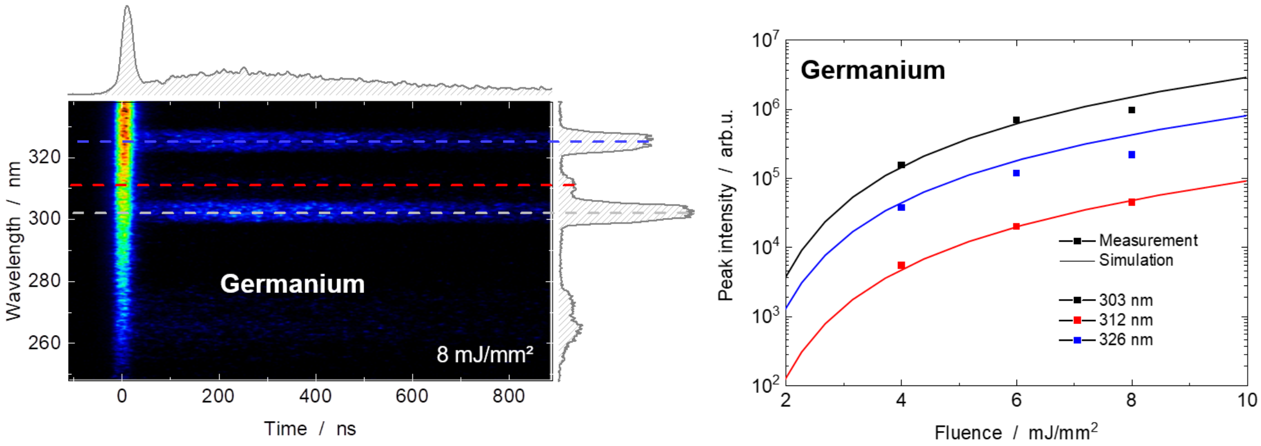

Fig. 1. Atomic line emissions of evaporated germanium atoms. (left) Spectrally and temporally resolved light emission during and after laser heating/evaporation. (right) Measured line emission intensity fitted with the line emission model from [1].

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Insights on laser-baked soot

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Despite the continuous efforts of the scientific community, Laser-induced Incandescence technique still presents some open questions and unresolved issues. Among them, the effect of rapid laser heating on the physical/chemical properties of the particles is far from being comprehensively explained. Changes in the particles internal structure and optical properties have been observed [1-3] but they are not taken into account in most of the LII models and often not accounted for in the interpretation of the data.

The aim of the present work is to clarify the effect of laser heating on soot optical properties by coupling optical and non-optical techniques. Soot particles were produced by a Nitrogen-quenched ethylene diffusion flame and heated at different temperatures by varying the laser fluence. Wavelength-resolved extinction measurements on laser-heated soot particles were performed in the visible spectral region. A significant variation of the extinction coefficient of the laser-heated soot particles compared to the extinction coefficient of the non-heated ones was observed. Such effect is particularly relevant below 550 nm and raises increasing the energy density of the laser. A similar trend was also observed for the absorption coefficient by using a seven-wavelengths Aethalometer. Nevertheless such behavior is in contrast with the common belief that laser heating of soot would promote a graphitization process and therefore a refractive index absorption function close to graphite [4] would be expected.

For a better understanding of this effect, laser-heated soot samples were analyzed by Raman spectroscopy, FT-IR spectroscopy and X-ray Diffraction. The particle size distribution of cold and heated soot was also measured.

Results suggest that different mechanisms, namely oxidation/fragmentation, vaporization etc. are probably responsible for the variation of laser-heated soot optical properties. However, further investigations are still necessary for a comprehensive understanding of the effect of intense laser radiation on soot particles and the impact on the interpretation of the LII data.

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Method and application of ambient black carbon mixing state measurements with the Single Particle Soot Photometer (SP2)

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The Single Particle Soot Photometer (SP2) simultaneously measures the incandescent and elastically-scattered light from individual particles crossing a continuous-wave, 1064 nm laser [1, 2]. With appropriate calibration SP2 incandescence and scattering signals are used to calculate black carbon (BC) mass-equivalent and overall optical diameters of individual particles, respectively. BC particles that are internally mixed with volatile or semi-volatile material present a challenge to the standard optical diameter measurement in the SP2, since the scattering signals of these particles are quickly distorted as they are heated in the SP2 laser and their non-BC material evaporates. To avoid this problem and optically size such mixed particles, a position sensitive scattering detector is employed in the instrument. Under certain assumptions, the information provided by the position sensitive detector and the scattering signals of non-evaporating particles can be used to reconstruct full scattering signals from the leading edges of scattering signals that are not yet perturbed by evaporation [3, 4]. This method is known as Leading Edge Only (LEO) fitting. LEO-fitted SP2 signals can be used to quantitatively characterize BC mixing state through combination of measured BC mass-equivalent and overall optical diameters (e.g. to calculate BC volume fractions, coating thicknesses under a core-shell approximation).

We will present an overview of the SP2 LEO-fit method for measuring ambient BC mixing state and its application, including validation and comparison of the procedure against both quantitative (aerosol particle mass analyzer [APM]-SP2 measurements) and qualitative (SP2 delay time method) measures of BC mixing state. The uncertainties in the method will be explored with reference to the underlying assumptions through comparison of BC-mass equivalent and optical diameters at the single particle level. Finally, we will present results from a field measurement campaign in Melpitz, Germany conducted in February – March 2017 that demonstrate both the ability and usefulness of the method.

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The morphology of soot aggregates generated in ethylene and propane inverse diffusion flames at different oxygen indexes

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The radiative properties of aggregated soot particles are strongly dependent on their morphology, besides wavelength and refractive index. This fact is also important to Laser Induced Incandescence of soot [1]. Inverse Diffusion Flames (IDF) [2] possess different thermal and chemical environments from normal diffusion flames (NDF) and have been less investigated in terms of the soot particle morphology. Few studies have focused on qualitative morphological analysis and primary particle sizes of soot sampled from ethylene/air IDFs. Jung et al. [3] showed that oxygen index (OI) influence the morphology of soot aggregates.

In this work, the morphology of soot aggregates in C_2H_4 and C_3H_8 IDFs at different OIs is investigated based on thermophoretic sampling at different height above the burner (HAB) and subsequent transmission electron microscopy (TEM) image analysis. Among the morphological parameters we report the fractal dimension (D_f), prefactor (k_f) and radius of gyration (R_g). Number (N_p), size ($d_{p,}$), polydispersity ($\sigma_{p,eo}$), and overlapping (C_{ov}) of monomers in aggregates are also calculated [4-6]. Based on these parameters, fractal aggregates were generated numerically using a modified Filippov et al. [7] tunable cluster-cluster aggregation algorithm. These simulated aggregates allow us to calculate not only additional morphological information such as aggregate volume (V_a), average projected area (A_p), surface area (S_a), anisotropy coefficient (A_{13}), and mobility radius (R_m), but also the thermal and radiative properties of such realistic soot aggregates. Aggregates generated from the C_3H_8-IDF were typically small. Meanwhile, a variety of aggregates were found in the C_2H_4-IDF. Examples of TEM images and the corresponding numerically generated aggregates are shown in Fig. 1.

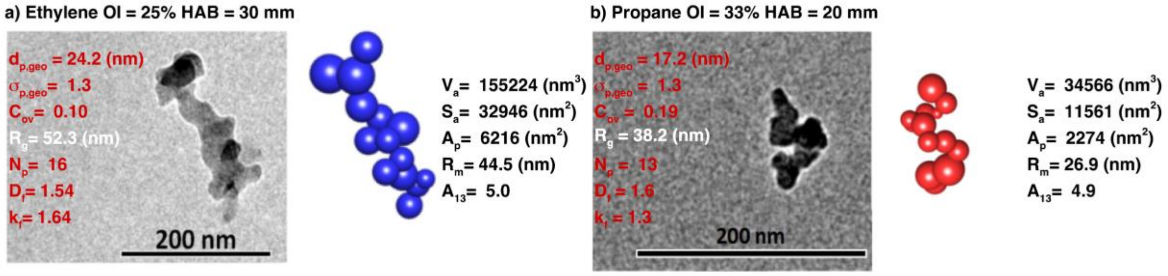

Fig. 1: TEM images for two aggregates sampled from an ethylene IDF, (a), and a propane IDF, (b). The numerically generated aggregates are also shown.

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Laser diagnostics for soot for high pressure CH_4-air diffusion flames

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Methane-air diffusion flames up to a pressure of 7 bar were studied using multiple laser diagnostic techniques. Laser-induced incandescence (LII), extinction, laser-induced fluorescence of polycyclic aromatic hydrocarbons (PAH LIF) and elastic light scattering (ELS) were the diagnostic techniques applied. LII combined with extinction could provide with 2D soot volume fraction distributions [1] while ELS when combined with LII or extinction could give information about the soot particle size [2]. PAH LIF gives information about soot formation by showing an indication of the precursors [3].

An Nd:YAG laser with a repetition rate of 10 Hz, operated at 532 nm is used for LII, PAH LIF and ELS. The laser beam was expanded and focused to a thin sheet using a combination of lenses and different ranges of fluences were selected appropriately for each diagnostic technique. A PI-MAX®4 ICCD camera was used as the detector. A short pass filter with a cut-off wavelength, 450 nm was used for LII to suppress interference from the flame luminosity. A notch filter at 532 nm (FWHM < 10 nm) along with a long pass 550 nm filter are used for PAH LIF measurements. An interference filter at 532 nm (FWHM = ±2.7 nm) along with a linear polarizer and in some cases a neutral density filter were used for ELS measurements. Extinction measurements were performed at 5 bar to calibrate LII signals into absolute soot volume fractions. A continuous wave diode laser operated at 850 nm was used for extinction measurements. Automatic background subtraction was enabled by ON-OFF modulations at 500 Hz of the CW laser output. Uncertainties caused in the gated LII signal acquisition due to the pressure dependence of the signal decay rates were corrected using a LII Model [4].

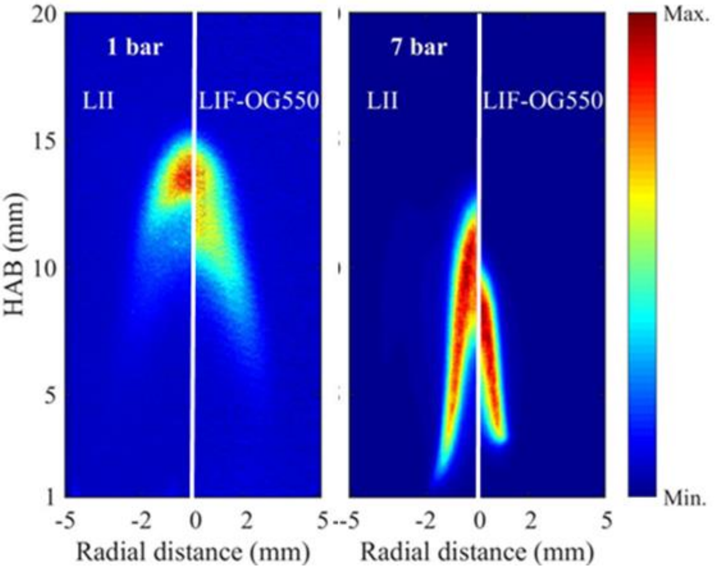

Fig 2: Combination of normalized LII (left) and PAH LIF (right) signal distributions at 1 bar and 7 bar pressures.

It was observed that soot volume fractions and concentrations of PAHs increases with increase in pressure, see Fig.1. It was also observed that PAHs starts to form at lower HABs compared to that of soot thus reinforcing the argument of PAH being considered as the precursors of soot.

Acknowledgements

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Temporally- and spectrally-resolved LII measurements on a standard flame using a streak-camera and multichannel PMT setup

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Two-color detection systems remain the most common in laser-induced incandescence (LII) applications. However, recently, three- [1,2] and four-channel [3] LII setups have been introduced, which allow the comparison of temperature decay curves derived from different two-color ratios, making the measurement technique more robust. In addition, using several detection wavelengths provides insight in the materials' spectroscopic properties. This is of interest, since LII has lately been extended to other materials than soot [4,5] for which often the optical properties are not yet known. In these cases, even higher spectral resolution is essential that can be achieved via detection with a streak-camera/spectrometer combination.

In this work, we compare the performance of a streak-camera/spectrometer system and a four-channel PMT setup with LII experiments on a standard flame (Gülder burner). Measurements were done using an Nd:YAG laser (1064 nm) at 42 mm height above burner with the burner operated at standard conditions (0.194 slm C_2H_4, 284 slm Air). LII signals were simultaneously detected with both setups (Fig 1).

The same spectroscopic model was applied to the LII signals recorded with both detection systems. The resulting temperature traces from both setups at various fluences and spectral ranges will be presented and differences discussed.

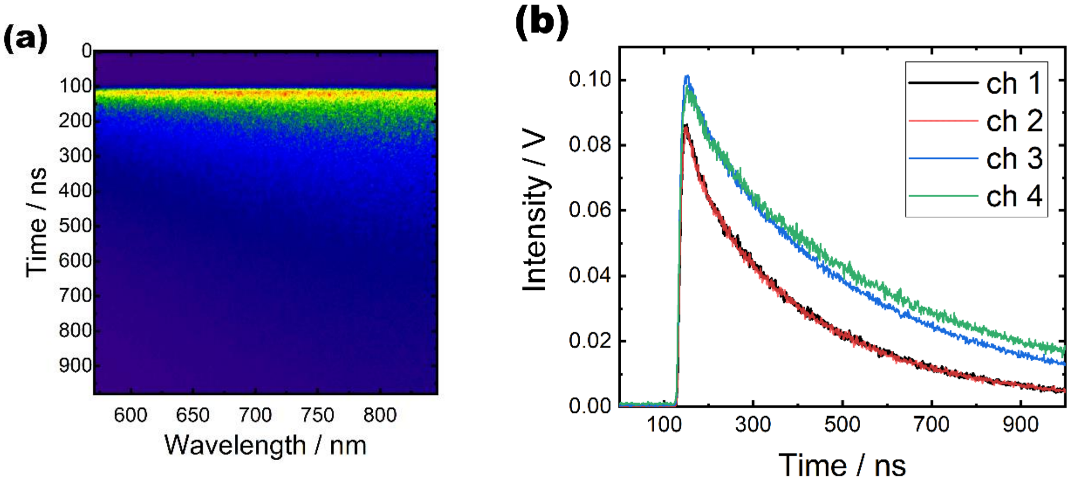

Fig. 1 (a) LII signal collected using a Streak Camera/Spectrometer; (b) LII signals collected using four-channel PMT setup (ch 1: 500 nm, ch 2: 500 nm, ch 3: 684 nm, ch 4: 797 nm).

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Investigation of soot formation in a novel diesel fuel burner

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Diesel engines are one of the major sources of particulate matter, which is widely recognized as a harmful pollutant [1]. However, emissions from diesel engines can be controlled by modifying the fuel chemical composition [2] for example by additive insertion. Fundamental studies of the effects of additives on soot formation are preferably performed on pre-vaporized fuels in order to separate the effects of additives from those of spray configuration and engine conditions. In the present work, a novel burner capable of complete pre-vaporization and stationary combustion of diesel in a diffusion flame was designed and built. The diesel burner was used with various types of diesel for the analysis of additive influence, i.e. a reference diesel and a baseline diesel with and without additives. For a better understanding of soot formation, a comprehensive characterization of the soot formed is required. Here, Laser-Induced Incandescence (LII) and Wide-Angle Light-Scattering (WALS) measurements were carried out for the determination of the soot volume fraction and soot aggregate size at different heights in the flame. While WALS measures radii of gyration of aggregates within a point-wise measurement volume, LII was employed in an imaging approach for a 2D-analysis of the soot volume fraction.

We will show results from LII and WALS measurements at different positions for different fuel types, and how fuel composition and fuel additives change the soot emissions during diesel combustion.

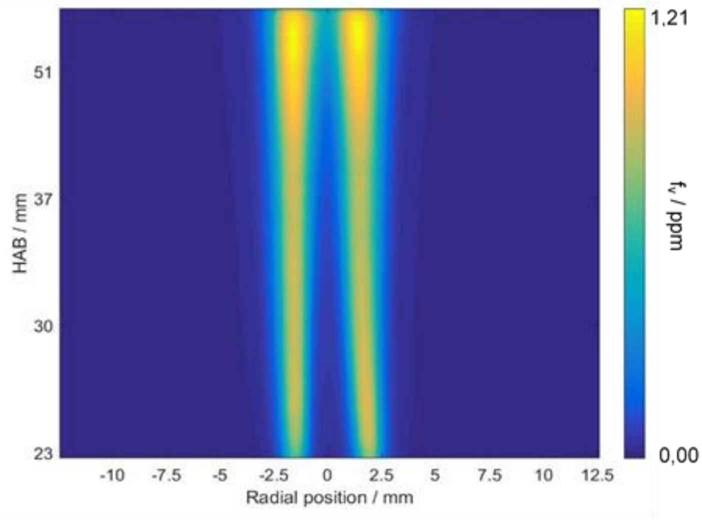

Fig. 1. LII mean image of the flame produced by combustion of the baseline diesel with the novel diesel-burner.

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LII measurements in a Confined Swirled Sooting Flame under Perfectly Premixed Rich Conditions

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A deep characterization of soot production in turbulent flames constitutes a central objective to reduce soot emissions of industrial devices. For this, soot production in laboratory-scale turbulent flames can be quantitatively investigated using the laser incandescence technique (LII) technique, a non-intrusive laser diagnostic that yields instantaneous soot volume fraction fields. Thanks to this, experimental investigations on turbulent jet diffusion flames and more recently on non-premixed swirled flames have greatly improved our understanding of soot production [1]. Nevertheless, the non-premixed situation is mainly controlled by the quality of mixing between fuel and oxidizer inducing local mixture inhomogeneities that complicate the understanding and the modeling of soot production. A review of the literature indicates that perfectly premixed rich sooting turbulent flames have been significantly less well documented. However such flames have considerable interest for different reasons. First, as fuel and oxidizer are initially mixed, they allow the characterization of turbulence effects on soot production without the influence of mixing. Second, perfectly premixed conditions represent an easier configuration for validation of numerical models. Finally, the investigation of soot production in this combustion mode is of relevance for the so-called Rich-Quench Lean configuration, a promising alternative for reduction of NOx emission in gas turbines.

These considerations have led to the design of a new experimental facility at EM2C laboratory designated as EM2Soot [2,3], allowing the stabilization of a perfectly premixed sooting flame for a wide range of equivalence ratios ($1.6<\phi<2.9$) and powers ($5\ \mathrm{kW}<P<30\ \mathrm{kW}$).

In this configuration, a high concentration of soot is detected via LII close to the quartz windows (Fig. 1), whereas light scattering measurements indicate that soot particles can be found everywhere in the chamber. Effects of flow rate, equivalence ratio and wall thermal conditions on soot production are examined both from direct images of the flame luminosity and LII measurements. These data can be used to guide the modeling of soot production in sooting rich premixed flames.

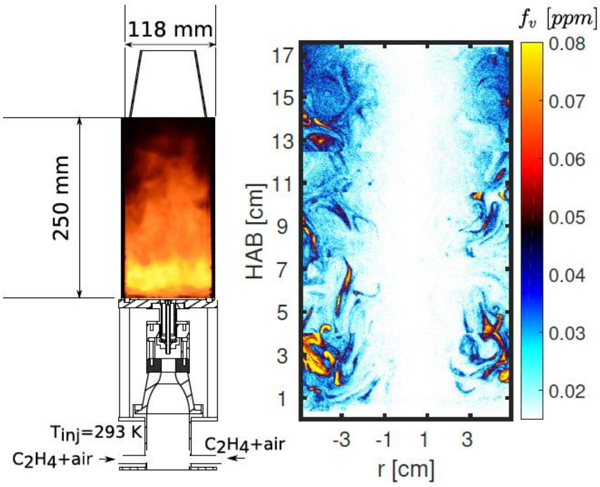

Fig. 1. Schematic of the EM2Soot burner (left) and 2-D mapping of instantaneous soot volume fraction by LII as a selective collage of images (right).

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Laser induced incandescence imaging in diffusion flames of liquid fuels relevant to biomass combustion

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The requirement for sustainable energy generation has led to an increase in the utilisation of biofuels and wood sources in combustion. It is important to investigate the formation of pollutants from using these alternative fuels, particularly the formation of aromatic hydrocarbons and soot. This study involves the use of liquid biofuels (Furfural, Eugenol, Anisole and a conventional hydrocarbon *n*-Decane for comparison) to represent components that are devolatilised as pyrolysis products during combustion of wood [1]. Diffusion flames of these fuels are stabilised on a simple wick burner and measurements are performed by LII planar imaging. Calibration of soot volume fraction was performed by making identical measurements in a flat flame burner in which extinction had been performed.

Time-resolved LII (TiReLII) profiles were also recorded. To do this, a correction for slight motions of the flame in successive images was required. This was done based on the position of the soot sheet identified at each height above the burner based on the location of the maximum LII signal.

Laser induced emission imaging was carried out at an excitation wavelengths of 532nm (in addition to 1064 nm) so that the incandescence signal could be subtracted to infer fluorescence from large aromatic soot precursors [2].

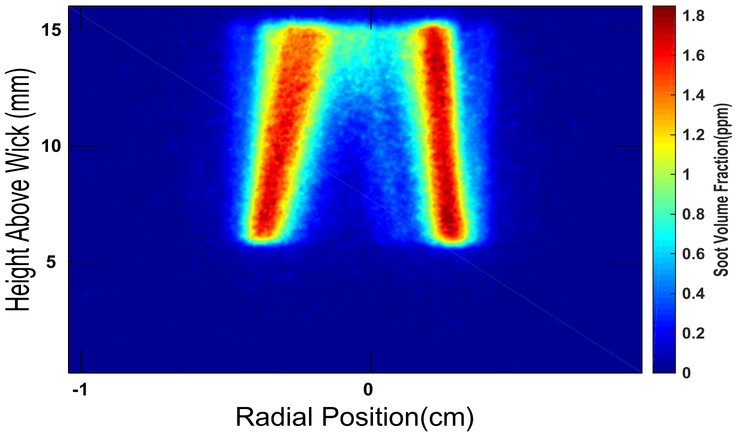

Fig. 1. Soot Volume Fraction in a furfural wick flame (average of 20 images)

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Laser induced incandescence (LII) using a long-pulsed fibre laser for *in-situ* study of soot in flames

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The requirement for methods to accurately measure soot volume fraction in challenging environments has become increasingly important due to progressively tighter emission standards and regulations. Laser induced Incandescence (LII) is one of the methods which can be used to measure soot concentration; it has high sensitivity and is non-intrusive in nature. For reasons of practicality and safety it may be attractive to use optical fibre for beam-delivery when performing LII in industrial test environments [1], which leads us to consider pulsed fibre lasers as a novel source for LII. Nevertheless, the temporal and spatial characteristics of such sources, as well as the repetition rate and the pulse energy, differ from those of Nd:YAG lasers generally used for LII. An investigation of the performance of LII conducted with a pulsed fibre laser has therefore been performed in a stable reference flame. One-dimensional images (see Figure 1) of soot volume fraction have been recorded at a range of flame conditions and heights above the burner and show good agreement with 'standard LII'. Fluence curves and time-resolved LII signals have also been studied with the fibre laser source. We discuss the advantages and limitations of this laser source for LII.

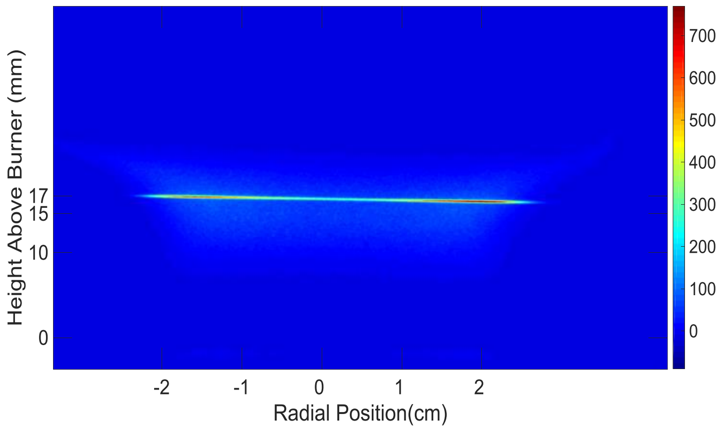

Fig. 1. Long Pulsed Laser LII Signal Image

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Two-dimensional LII for in-situ soot characterization of propane flames and influence of additives in a 100 kW oxy-fuel furnace

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Laser-induced incandescence (LII) and extinction measurements were successfully applied for in-situ soot characterization in a 100 kW_{th} down-fired oxy-fuel test furnace. A Nd:YAG laser operated in its fundamental mode at 1064 nm was used for LII helping thereby avoiding signal interferences from polycyclic aromatic hydrocarbons (PAHs) [1]. A laser fluence of 0.8J/cm^2 was set for the LII measurements based on test measurements on ethylene-air flames (ϕ = 2.3) on a McKenna burner. A PI-MAX®4 ICCD camera was used as the detector. A short pass filter with a cut-off wavelength 450 nm was used for LII to suppress the interference with background flame luminosity. Moreover, background flame radiation was captured 5 µs before the LII signal for automatic background subtraction. LII signals were calibrated to soot volume fractions, f_v, using in-situ extinction [2] in the same spatial regions of the furnace. A continuous wave diode laser operated at 808 nm was used for performing extinction measurements. Automatic background subtraction was enabled by ON-OFF modulations at 500 Hz of the CW laser output. Moreover, selection of this wavelength helped in avoiding laser absorption by PAHs [3].

Two-dimensional single shot LII signals from soot show strong spatial variations as well as local temporal variations, see Fig.1. The study primarily focused on non-premixed propane flames in an oxy-fuel mode with various oxygen concentrations in the oxidant. Moreover, the influence of additives was studied for a few measurement cases for both oxy-fuel and air environments. A decrease in f_v was observed for additives SO_2 and NO for oxy-fuel conditions, while a decrease was found for air-fed flames. Additio-nally, water injection in the air-fed flames showed large decrease in f_v, while water with additional KCl dissolved in it reduced f_v somewhat more.

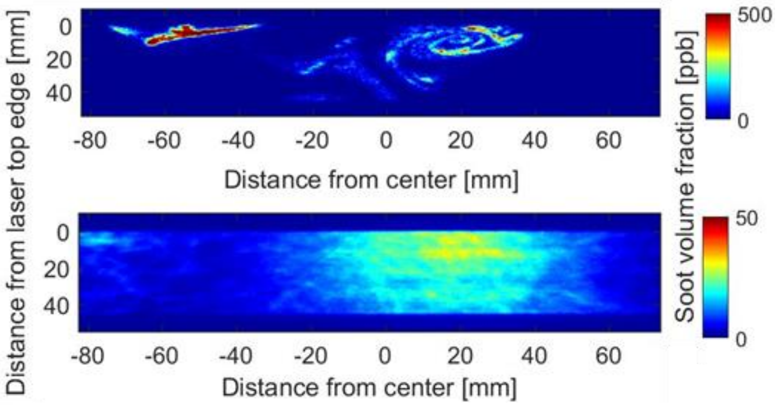

Fig. 1. Single shot (top) and averaged (bottom) soot volume fraction distribution for oxy-fuel case with 40 % O_2.

Acknowledgements
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What is hiding in the intensity scaling factor and what can be gained from analyzing its temporal variation?

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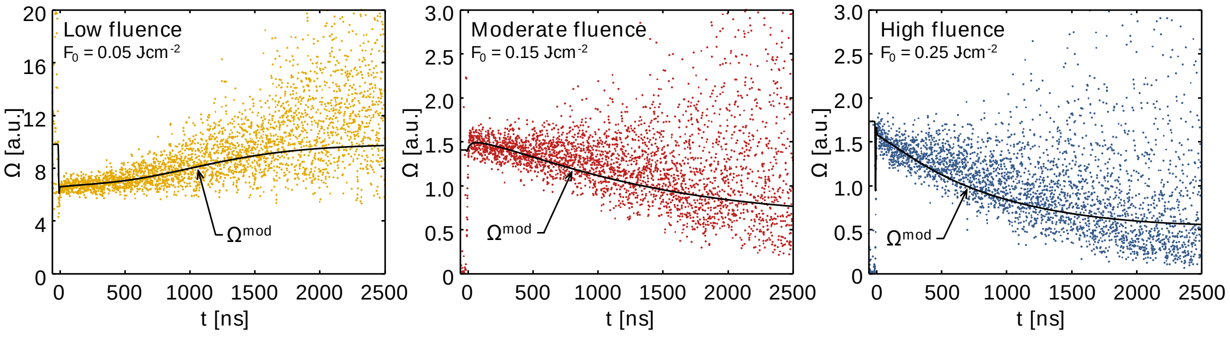
Time-resolved laser-induced incandescence (TiRe-LII) researchers have long used a scaling factor to relate the incandescence from laser-heated nanoparticles to a detector voltage [1-3]. The definition and terminology has varied widely throughout the literature and has encompassed any number of other parameters including the volume fraction, detector gain, and optical geometry. During inference, researchers have also noted temporal changes in these parameters that cannot be explained using current models [2,3]. In this work, we examine these temporal changes and develop a framework for identifying these effects and how they might impact analysis of LII data. We adopt the naïve approach of incorporating these effects into a single parameter called the *intensity scaling factor* (ISF), Ω, implicitly defined as

$$S_\lambda = \Omega(t) \cdot \eta_\lambda \cdot C_{\mathrm{abs},\lambda}(d_\mathrm{p}) \cdot I_{\mathrm{b},\lambda}\left[T_\mathrm{p}(d_\mathrm{p},t)\right], \quad (1)$$

where S_λ, is the LII detector signal, $C_{\mathrm{abs},\lambda}$ is the nanoparticle absorption cross section, η_λ is the calibration coefficient, and $I_{\mathrm{b},\lambda}$ is the spectral blackbody intensity.

In examining the temporal changes in the inferred value of the ISF, we consider five effects: (i) sublimation; (ii) soot annealing, using a simplified model; (iii) polydispersity, as it directly affects the ISF; (iv) background luminescence, for the case where the data is not properly corrected to account for this effect; and (v) the detector response function, which is modeled as a Gaussian temporal response. We examine how each of these effects can result in temporal changes in the ISF over different time scales. Finally, the predicted changes in the ISF are compared to experimental observations. Figure 1 shows these results for low, moderate, and high fluences relative to the regimes defined in Ref. [4]. Many of the characteristics in the experimental curves are reflected by the proposed ISF model.



(a) (b) (c)

Fig. 1. Experimentally-inferred (points) and predicted (line) ISF curves for a range of fluences including the five effects noted in the text.

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Predicting the heat of vaporization of iron at high temperatures using TiRe-LII and Bayesian model selection

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Over the last two decades, considerable focus has been placed on improving the fidelity of TiRe-LII heating and cooling models, with the objective of improving the robustness of inferred aerosol parameters [1,2]. While simple models fail to capture all the salient features of experimental cooling curves, more elaborate models that account for such features also introduce additional degrees-of-freedom and complex functional forms, which increase the uncertainty in recovered parameters and make them susceptible to over-tuning. Therefore, identifying the appropriate level of model complexity is one of the most important questions facing LII researchers.

Bayesian model selection is a promising tool for fulfilling this task [3]. In this approach, competing models are evaluated in terms of fit to the measurement signal as well as their complexity, accounting for measurement noise and both the number and the degree of prior information available about the model parameters. The procedure is based on the calculation of a Bayes factor [3], which quantifies the relative probability of two models in a universal way. We further decompose the Bayes factor into the relative fit, ΔF, which accounts for agreement between estimates and the data, and relative measurement credence, ΔC, which penalizes parameters that are difficult to infer from the measurements [4]. The Bayes factor itself obeys a probability density due to noise in the data.

We demonstrate the Bayesian approach to model selection using simulated and experimental TiRe-LII data for iron [5]. Figure 1 illustrates the significance of the evaporation submodel in the analysis of iron data, showing ΔF, ΔC, and the Bayes factor as a function of peak temperature. There is a marked decline in the Bayes factor when the evaporation model becomes significant. We also test the suitability of Román’s equation, a universal expression that predicts the latent heat of vaporization. Our results show that this equation best describes the latent heat of vaporization of iron over the large temperature range typical of TiRe-LII data. More generally, this procedure illustrates how TiRe-LII can be used to determine the fundamental thermophysical properties of nanoparticles, including soot, and introduces a method to answer ongoing questions about the disputed properties of soot.

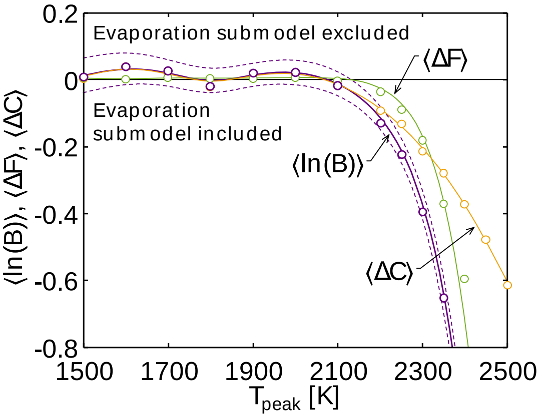

Fig. 1. Trends in the logarithm of the Bayes factor, relative fit, and relative measurement credence as a function of peak temperature. Values less than zero indicate a preference for including the evaporation submodel.

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Neutral bremsstrahlung emission in laser-induced incandescence experiments on soot and silver nanoparticles

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Recent experiments of low-fluence pulsed time-resolved laser-induced incandescence (TiRe-LII) on a soot-laden aerosol using an excitation wavelength of 532 nm reveal an anomalous signal starting shortly after the start of the laser pulse (Fig. 1a and 1b) that decays during the laser pulse at a rate independent of the laser fluence. Such characteristics cannot be explained purely by nanoparticle cooling, and indicate the presence of another emission phenomenon. These observations have also led to reconsideration of published TiRe-LII measurements on silver nanoparticles (Fig. 1c) [1]. The insensitivity of the peak temperature to the laser fluence, the unexpected rapid temperature decay, and higher absorption efficiency relative to Mie and Drude theory suggests that the observed emission is unlikely to be incandescence and may be another example of this unexplained phenomenon.

The current work investigates the possibility that neutral bremsstrahlung emission is responsible for the anomalous signal. In the absence of significant incandescence, neutral bremsstrahlung is a likely candidate for anomalous emission and enhanced absorption. Electrons originate from thermionic emission and thermally-assisted multi-photoemission from the nanoparticle and induce a plasma. As the electrons are emitted, a positive charge builds up in the nanoparticle, stopping further electron release. This limits electron emission to the beginning of the laser pulse, and the bremsstrahlung emission will decay before the peak laser fluence as the electrons diffuse through the gases. This will result in nearly identical decay times in the anomalous signals for Ag and soot, an observation noted in experiments.

This work presents a model for this phenomenon that includes electron emission from the nanoparticle during the laser pulse and neutral bremsstrahlung emission as the electrons diffuse through the gas phase. Modeled signals are compared with experiments, including their sensitivity to different bath gases (Ar, N_2, and CO_2) [1]. The results indicate that, in some low fluence experiments, neutral bremsstrahlung could significantly contribute to the observed LII signal.

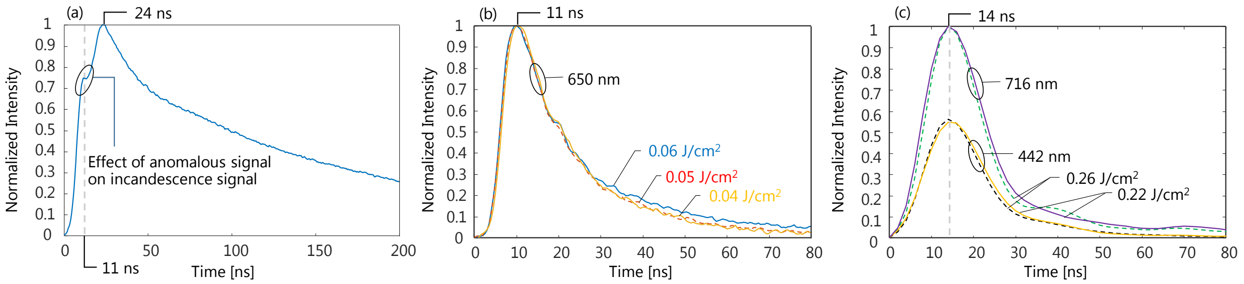

Fig. 1. LII signals (a) for soot at a fluence of 0.1 J/cm^2, (b) for soot at very low fluences (only the anomalous signal), and (c) for silver at two colors. All signals are normalized to the peak of the 716 nm channel.

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Laser-induced incandescence on metallic nanoparticles: Investigating effect of plasma thermal bremsstrahlung emission on peak temperature pyrometry inference

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Current time-resolved laser-induced incandescence (TiRe-LII) models cannot explain several phenomena in the experimental data, including: (*i*) “anomalous cooling”, particularly pronounced in measurements on low-temperature aerosols, in which the nanoparticles appear to cool faster than can be explained via conduction and sublimation/evaporation submodels [1,2]; (*ii*) a higher absorption cross-section of some metallic nanoparticles at the laser wavelength compared to Mie/Drude predictions [2]; and (*iii*) a temporary augmentation of extinction during and shortly after the laser pulse as a function of laser-fluence in combined LII/line-of-sight-absorption (LOSA) measurements [3].

Recent experiments suggest that these discrepancies may arise from a laser-induced microplasma that envelops the nanoparticle [4,5]. Excessive absorption can be justified by the inverse bremsstrahlung process of the plasma. The plasma also emits blackbody radiation according to the spontaneous plasma temperature, which corrupts the spectral incandescence emitted by the nanoparticle while the plasma dissipates; this may account for the anomalous cooling phenomenon, cf. Fig. 1.

This work presents a theoretical underpinning for laser-induced plasma formation under LII measurement conditions, and explores how this plasma may affect TiRe-LII measurements on Fe, Mo, molten Si, Ag and W nanoparticles. At fluences greater than 0.8 J/cm^2, the absorption cross-section of laser-energized nanoparticles is enhanced due to inverse bremsstrahlung absorption, and bremsstrahlung emission causes overestimation of the nanoparticle temperature due to the corruption of the incandescence signal. Under these conditions, neutral bremsstrahlung emission is more prevalent than electron-ion bremsstrahlung due to the weak nature of the induced plasma. Nevertheless, the current model does not predict laser-induced plasma phenomena below ~0.5 J/cm^2, typical of low-fluence TiRe-LII measurements.

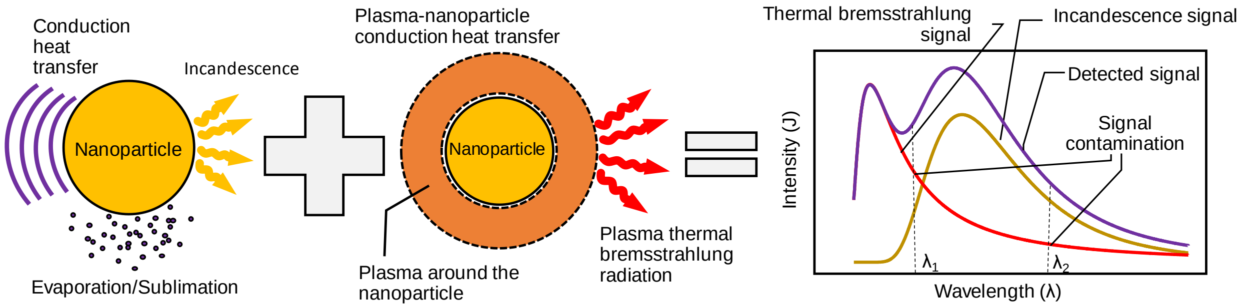

Fig. 1. Plasma bremsstrahlung corrupting LII incandescence signal.

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Laser-based experimental investigation on soot evolution during coal combustion in O_2/N_2 and O_2/CO_2 conditions

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Laser-induced incandescence (LII) is introduced as a valuable technique for the characterization of soot particles. Based on the spatially resolved measurement using this technique, soot evolution process has been better understood, but mostly in the gaseous hydrocarbon flames. The difficulties of arrangement for solid fuel combustion and presence of other kinds of carbonaceous particles in the flames make the application of LII more complicated in solid fuel flames. Only few investigations can be found in [1, 2]. In the current work, the laser diagnostics, including LII, laser elastic scattering (LES), and laser-induced fluorescence (LIF), were applied to characterize the particles as well as polycyclic aromatic hydrocarbons (PAHs) spatially. Based on the derived dataset, an attempt was made to explore the effect of oxygen concentration on the soot evolution during coal combustion in O_2/N_2 and O_2/CO_2 conditions.

The experiments were performed in a Hencken type entrained-flow reactor. Thin round stainless steel tubes were inserted in a square honeycomb ceramic plate and through which small diffusion flames from methane were established near the burner exit to generate hot flue gas. The pulverized coal particles were introduced into the hot flue gas from the center, and the particle jet flame can be seen in Fig.1. The temperature and composition of the flue gas was kept constant while the oxygen concentration of the gas carrying particles was varied from 0 to 100%.

The measured results showed that the flames became shorter while the particle temperature increased with the oxygen concentration. The scattering signal became weaker with height but showed slight increases nearby the burner exit and the sooting region in some cases. PAH was mainly distributed in the region below the region with high LII signal, similar to the diffusion flame from gas fuel. Soot volume fraction was calibrated in an ethylene/air diffusion flame established at the same position of coal jet flame. It was indicated that the peak soot volume fraction was around 0.5-2.5 ppm and increased slightly followed by a steep decrease as the oxygen concentration increased.

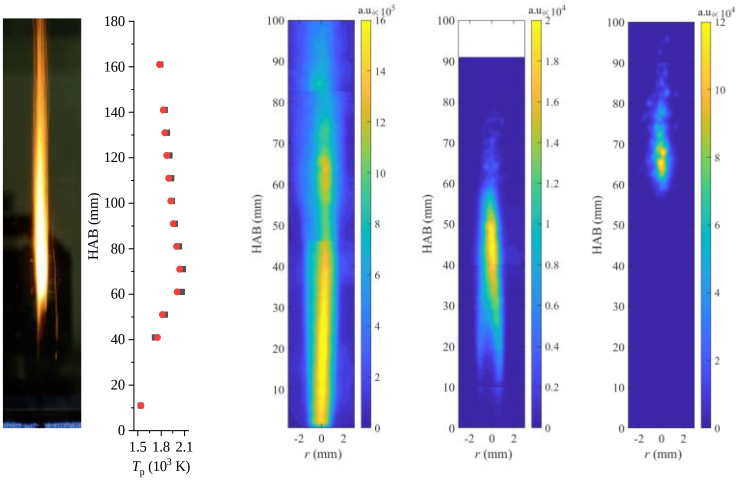

Fig. 1 Results of visible flame image, temperature along the flame axis, Mie scattering, PAH-LIF, soot-LII (from left to right) in the case of $20\%O_2/80\%N_2$. The height range of the visible flame image is the same with that of the temperature profile.

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Comparison of continuous wave and pulsed LII measurements of black carbon mass at atmospherically-relevant concentration levels

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Two types of LII systems are currently used to measure particulate black carbon (BC) mass in the atmosphere: continuous wave LII (employed most commonly in the single particle soot photometer, SP2, DMT Inc., US) and pulsed LII (for which several different custom-built and commercial versions exist) [1]. Pulsed LII is less widely used than the SP2 in the atmospheric science community. A few published studies have compared pulsed LII in terms of BC mass concentration with the techniques widely used in atmospheric science, such as the SP2, and other techniques, such as EC/OC analyzer and aethalometer [2-4]. There is particular interest in comparing pulsed LII with the SP2 since these techniques share the same LII operating principle. Currently the LII-300 (Artium Inc., US) is the only commercialized pulsed LII instrument. It employs a standard two-color auto compensating (AC) mode as well as a high sensitivity (HS) mode, recording incandescence signal only in a single channel. The HS mode nominally allows the instrument to measure the low concentrations of BC typically found in the atmosphere ($< \sim 5\ \mu g\ m^{-3}$). This study aimed to evaluate the accuracy of the AC and HS modes of the LII-300 at atmospherically-relevant concentrations via comparison against reference BC mass concentration measurements in laboratory and field experiments. We focused on the following major research questions: What is the limit of detection for the AC and HS modes? Do the measurements from each mode agree in the region of overlap? Do measured BC mass concentrations in each mode agree with reference measurements (SP2 measurements in the field and APM [aerosol particle mass analyzer]-referenced SP2 measurements in the laboratory)? Does the level of disagreement or agreement depend on BC type and/or BC mass concentration? The field measurements were performed in Bologna, Italy in July 2017 and involved a second pulsed LII system, custom built at CNR-ICMATE, Italy. BC mass concentrations measured by the two pulsed LII systems correlated well with each other as well as with those measured by an SP2 and a MAAP (multi-angle absorption photometer). However, the absolute scaling of the two pulsed LII systems differed, with BC mass concentrations measured by the LII-300 and the CNR-LII differing from those measured by SP2 by average factors of 0.6 and 1.9, respectively. Possible reasons for these discrepancies will be explored.

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