Fifth International Workshop on Laser Induced Incandescence

Palais des Congrès, Le Touquet, France
May 8-11, 2012

PC2A

Université Lille1
Sciences et Technologies
UFR de Chimie

The 5th International LII Workshop provides a forum for open high-level discussion on the understanding of LII diagnostics and to foster relationships in joint research. Laser-induced incandescence (LII) has proven to be a powerful tool for particle concentration and particle size measurements in combustion, particle synthesis, as well as in environmental applications. However, different experimental approaches and data evaluation techniques exist which, while demonstrating the complexity of the physical processes involved in LII and the need for further research, has also somewhat hampered the acceptance by industry of LII as a measurement standard. In order to strengthen the community and to explore the development of a series of best practices for LII modeling, experiments, and data interpretation, a series of workshops was initiated in Duisburg, Germany (2005), followed by workshops in Bad Herrenalb, Germany (2006), Ottawa, Canada (2008) and Varenna, Italy (2010).

The 5th International LII Workshop will be held May 9–11, 2012 in Le Touquet, France

Organizers

Pascale Desgroux, Sébastien Batut, Eric Therssen ; PC2A, CNRS/Université Lille1,
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We acknowledge the financial support of :

Program

Tuesday, May 08
Arrival, Check-in, Registration 17:30-19:30 at Palais des Congrès
General information

Wednesday, May 09

8:00 Registration
8:45 Welcome

Session 1: Experiment and modelling for LII fundamental knowledge

Experimental

9:00 Pulsed laser heating of differently aged soot probed using LII and LES
Nils-Erik Olofsson, Jonathan Johnsson, Henrik Bladh, Per-Erik Bengtsson
Lund University

9:20 Experimental setup to study soot sublimation as typically occurring in high fluence LII
Klaus Peter Geigle, Gregor Gebel, Markus Köhler
German Aerospace Centre (DLR), Stuttgart

Modelling

9:40 Influence of soot aggregate structure on particle sizing using LII
Jonathan Johnsson, Henrik Bladh, Nils-Erik Olofsson, Per-Erik Bengtsson
Lund University

10:00 Extending Time-Resolved LII to Metal Nanoparticles: Simulating the Thermal Accommodation Coefficient
K. J. Daun, J. T. Titantah, M. Karttunen and T. A. Sipkens
University of Waterloo, University of Western Ontario

Coffee break: 10:20-11:00 (registration continuation)

Session 2: ELS fundamentals

11:00 A method for inferring the soot size distribution by Static Light Scattering : Application to the CAST soot generator
Jérôme Yon, Chloé Caumont-Prim, Alexis Coppalle, Kuan Fang Ren
CORIA, University and INSA Rouen

11:20 Recent applications of the WALS-technique
Hergen Oltmann, Stefan Will
Universität Bremen, Universität Erlangen-Nürnberg

11:40 **Poster advertising I**

12:30-14:00: Lunch

Session 3: Combined methods for better knowledge of soot size

14:00 Soot primary particle sizing in turbulent flames via combined LII and Elastic Light Scattering
Brian Crosland, Matthew Johnson, Kevin Thomson
Carleton University, NRC Ottawa

14:20 Defining a measurement strategy for 2D soot particle size imaging through detailed LII signal-decay analysis
Emre Cenker, Gilles Bruneaux, Thomas Dreier, Christof Schulz
IFP En, Rueil-Malmaison, IVG and CENIDE, University of Duisburg-Essen

14:40 **Poster advertising II** (follow up)

Free afternoon around 15:00

Poster session (with refreshment)

20:00- 22:30

Thursday, May 10

8:50 Opening of the workshop discussion (objectives)

9:00 **Discussion on LII modelling**

10:30-11:00: coffee break

Session 4: Experiments for particles properties fundamental knowledge

11:00 Effects of particle coatings on laser induced incandescence
Ray Bambha, Paul Schrader, Mark Dansson, Hope Michelsen
SANDIA

11:20 Determination of the dimensionless extinction coefficient for soot generated by a PMMA flame
Damien Hebert, Alexis Coppalle, Jérôme Yon and Martine Talbaut
CORIA, University and INSA Rouen

Session 5: Combined methods

11:40 A combined laser induced incandescence, aerosol mass spectrometry, and scanning mobility particle sizing study of non-premixed ethylene flames
Scott Skeen, Paul Schrader, Kevin Wilson, Nils Hansen, Hope Michelsen
SANDIA, Lawrence Berkeley National Lab

12:00 Soot particles detection by LIBS and LII analysis
Francesca Migliorini, Silvia Maffi, Silvana De Iuliis, Giorgio Zizak
CNR-IENI, Milano

12:30-14:00: lunch

14:00 **Discussion on Experimental LII**

Discussion about the LII workshop follow up

Poster session (and refreshments)
16:00-18:00

19:00 banquet

Friday, May 11
Session 6: applications

8:30 LII and one-wavelength Aethalometer measurements of particulate matter in different environments
Silvana De Iuliis, Silvia Maffi, Francesca Migliorini, Giorgio Zizak
CNR-IENI, Milano

8:50 Time-resolved Laser induced incandescence measurement for a combustion field of the 0.5 kg-coal/h pulverized coal jet burner
Jun Hayashi, Nozomu Hashimoto, Noriaki Nakatsuka, Hirofumi Tsuji, Hiroaki Watanabe, Hisao Makino, Fumiteru Akamatsu
Osaka University

9:15 **Discussion on combined and emerging approaches**

10:45-11:15: Coffee break

11:15 **Summary and conclusion of the workshop, hot topics, discussion about a next workshop**

12:15 buffet

List of oral presentations

(alphabetic order first author)

Effects of particle coatings on laser induced incandescence
Ray Bambha, Paul Schrader, Mark Dansson, Hope Michelsen
SANDIA

Defining a measurement strategy for 2D soot particle size imaging through detailed LII signal-decay analysis
Emre. Cenker, Gilles Bruneaux, Thomas Dreier, Christof Schulz
IFP En, Rueil-Malmaison, IVG and CENIDE, University of Duisburg-Essen

Soot primary particle sizing in turbulent flames via combined LII and Elastic Light Scattering
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LII and one-wavelength Aethalometer measurements of particulate matter in different environments
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An experimental setup to study soot sublimation as typically occurring in high fluence LII
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Determination of the dimensionless extinction coefficient for soot generated by a PMMA flame
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Influence of soot aggregate structure on particle sizing using laser-induced incandescence
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University of LUND

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A method for inferring the soot size distribution by Static Light Scattering :
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Effects of particle coatings on laser induced incandescence

Ray Bambha, Paul Schrader, Mark Dansson, Hope Michelsen

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In exhaust plumes and under some combustor conditions soot particles are often coated with unburned fuel, sulfuric acid, water, ash, and other combustion by-products.[1,2] Diesel particles, for example, can be comprised of as much as 50% volatile compounds.[3] These coatings can have an effect on particle optical properties and can thus have an influence on optical diagnostics applied to coated particles. The effects of particle coatings therefore need to be fully understood in order to apply optical diagnostics under a wide range of conditions.

We have compared time-resolved laser induced incandescence (LII) measurements on uncoated soot generated in a coflow diffusion flame with LII measurements on heavily coated soot generated in a fuel-rich premixed flame. Soot was extracted and cooled from both flames, and a thermodenuder was used to vary the coating on the particles extracted from the premixed flame. A scanning mobility particle sizer (SMPS) was used to monitor aggregate sizes from the two flames, and transmission electron micrography (TEM) was used to characterize particle morphologies. The results demonstrate striking differences in LII temporal evolution and dependence on laser fluence between coated and uncoated particles. These results can be understood in the context of particle energy balance during heating and cooling and are consistent with predictions based on an LII model that includes a heavy organic coating.

References

(1) Kittelson, D. B. *J. Aerosol Sci.* **1998**, *29,* 575-588.

(2) Lighty, J. S.; Veranth, J. M.; Sarofim, A. F. *J. Air Waste Manage. Assoc.* **2000**, *50,* 1565-1618.

(3) Witze, P. O.; Gershenzon, M.; Michelsen, H. A. *Proc. SAE* **2005**, SAE Paper no. 2005-01-3791.

Defining a measurement strategy for 2D soot particle size imaging through detailed LII signal-decay analysis

E. Cenker[1,2], G. Bruneaux[1], T. Dreier[2], C. Schulz[2]

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[2]*IVG and CENIDE, University of Duisburg-Essen, Duisburg, Germany*

A combination of two-color soot pyrometry imaging, two-color time-resolved LII (TiRe-LII), Laser Extinction Method (LEM) and Transmission Electron Microscopy (TEM) of soot samples is used to define a strategy for two-dimensional imaging of soot particle size distributions. TiRe-LII is carried out both by single point measurements and 2D imaging, where LII signal-decay is determined for each pixel through time-gate-sweeping of the camera gate relative to the laser pulse. Experiments are carried out on an atmospheric laminar ethylene/air diffusion flame from a Santoro burner with a 1064-nm laser sheet operated in the low-fluence regime. For flame temperature measurements, two-color pyrometry images are tomographically inverted. The resulting temperature fields are used as input for the evaluation of the primary particle size from the local LII decay curves using the LIISim model.

It was found that the LII signal shows different decay properties at different delays after laser heating. As the particles cool down towards ambient temperature, calculated decay constants increase. For closer inspection, the TiRe-LII signal is divided into several 100-ns long segments and individual particle sizes are calculated for each segment by two-color LIISim curve-fitting. As the time-window is shifted further away from the laser, larger particle sizes are calculated. For a delay of 700 ns between two segments, the calculated particle size difference is greater than 12 nm. This variation is attributed to the polydisperse nature of the particle size distribution in the region of interest where small particles cool down to ambient temperature within a few hundred nanoseconds and their contribution to the detected LII signal fades out.

The dependence of the predicted particle sizes on the boundary conditions imposed for the simulation, such as ambient temperature, agglomeration, and accommodation coefficients are also quantitatively investigated. For validation of the evaluated particle sizes and uncertainty analysis, particles are sampled at different locations in the flame above the burner head via thermophoretic sampling on TEM grids. Primary particle sizes and dispersion are derived from TEM micrographs.

For the next step, these techniques will be applied in a high-pressure burner and a high-pressure spray vessel. In light of the time-segmented decay analysis, an optimized gate positioning for 2D-LII and a comprehensive simulation model strategy will be determined.

Soot Primary Particle Sizing in Turbulent Flames via Combined LII & Elastic Light Scattering (ELS)

Brian Crosland[1], Matthew Johnson[1], Kevin Thomson[2]

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Laser Induced Incandescence (LII) is often promoted as a real-time, in situ, spatially resolved optical method capable of measuring soot volume fraction f_v and soot primary particle diameter d_p through analysis of time-resolved incandescence data and the application of a soot cooling model. However, a key parameter of any soot cooling model is the local gas temperature since it drives the cooling process. While it is possible to spatially map a flame gas temperature field for steady flames, measurement in unsteady flames would require simultaneous measurement of the unheated gas temperature and the subsequent LII cooling curve to reliably determine soot primary particle diameter, thus limiting the applicability of LII soot sizing in unsteady flames.

The current work aims to avoid the need for gas temperature measurement and time-resolved LII by instead obtaining d_p as well as the soot radius of gyration R_g via combined LII and ELS. It has been shown that the absolute magnitude of backward scattering from soot aggregates is mostly independent of aggregate size, instead depending on primary particle diameter and soot volume fraction[1] while the ratio of a forward and backward scattering measurement can be used to measure R_g[2].

A 532 nm pulsed Nd:YAG laser operating at the cusp of the plateau regime is used to produce both LII emission and elastic light scattering. A single optical collection tube collects both LII emission and scattering at an angle of 150°. The collected signal is subsequently separated into three wavelength bands (two for auto-compensating LII and one for ELS), which are detected using photomultiplier tubes (PMTs). The PMT output is amplified and processed by a gated integrator to produce an average measurement during the gate width. ELS at an angle of 30° is collected by a second optical collection tube coupled to a fourth PMT.

Ensemble-averaged measurements made on the centerline of a buoyancy-driven non-premixed turbulent flame burning a mixture of mostly methane indicate d_p on the order of 20-30 nm and R_g from 60-100 nm for mean soot concentrations up to 0.15 ppm. Flame radiation has a negligible contribution to both the LII and ELS signals, as does secondary light scattering within the combustion enclosure. The bias uncertainty in the measurements is driven by the uncertainty on the soot refractive index absorption function $E(m_\lambda)$ and the calibration of the neutral density filters used to attenuate the PMTs used for scattering measurements, while the precision uncertainty is limited by photon shot noise.

[1] Teng, Y., & Koylu, U. O. (2006). Optical sizing of aggregated combustion particles: computational development of a two-angle laser scattering technique. *Applied optics*, 45(18), 4396-403.

[2] De Iuliis, S., Cignoli, F., Benecchi, S., & Zizak, G. (1998). Determination of soot parameters by a two-angle scattering-extinction technique in an ethylene diffusion flame. *Applied optics*, 37(33), 7865-74.

Extending Time-Resolved LII to Metal Nanoparticles: Simulating the Thermal Accommodation Coefficient

K. J. Daun[a*], J. T. Titantah[b], M. Karttunen[a] and T. A. Sipkens[a]

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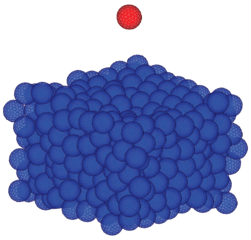
There is growing interest in adapting time-resolved laser-induced incandescence (TiRe-LII) to size metal nanoparticles, owing to their emerging applications in materials science. Extending TiRe-LII to new aerosols requires a model for the heat transfer between the laser-energized nanoparticles and the surrounding gas. Unfortunately, the thermal accommodation coefficient, α, which defines the energy transferred when a gas molecule scatters from the particle surface, is rarely available. This parameter can sometimes be obtained from LII measurements made on a reference aerosol sized using electron micrography, but this process is notoriously time-consuming, and thermophoretic sampling of metal nanoparticles is often problematic. These challenges have precluded interpretation of data from several pioneering TiRe-LII studies on metal nanoparticles, including one by Murakami et al. [1] that intended to determine how the bath gas influences the growth of molybdenum nanoparticles formed through laser-induced photolysis of $Mo(CO)_6$.

Alternatively, it is sometimes possible to estimate α using molecular dynamics (MD). In this technique, a pairwise potential between the gas molecule and metal atoms is derived from ab initio (generalized gradient approximations of density functional theory, GGA-DFT) calculations of the gas/surface potential. The potentials then differentiated to obtain forces, and Newton's equations of motion are time-integrated to obtain atomic trajectories during a gas/surface scattering event. Finally, α is found through Monte Carlo integration over all incident gas molecular trajectories.


MD simulation of an argon molecule scattering from a laser-energized iron nanoparticle

This approach was initially used to characterize α between soot and various gases, and is presently being extended to metal nanoparticles. Preliminary results show that MD-derived accommodation coefficients are highly sensitive to the potential well depth. Unfortunately, a well-known limitation of GGA-DFT is that they cannot describe the long-range electron correlations responsible for van der Waals (vdW) forces, which contribute to the potential well. While the Ni/Ar interaction is dominated by a strong Casimir force, vdW forces are thought to play a major role in other systems. Accordingly, true accommodation coefficients are probably larger compared to ones found using ab initio derived gas-surface potentials with no vdW correction. Current research is focused on identifying an appropriate heuristic correction that can account for the dispersive forces.

Preliminary thermal accommodation coefficients for metal nanoparticles

	α_{MD}	α_{exp}
Ni/Ar	0.20±0.02	
Fe/He	0.07±0.01	0.01 [2]
Fe/Ar	0.04±0.01	0.1 [2], 0.13 [3]
Mo/He	0.006±0.002	
Mo/Ar	0.04±0.01	

[1] Y. Murakami, T. Sugatani, Y. Nosaka, J. Phys. Chem., 109 (2005) 8994.
[2] A. Eremin, E. Gurentsov, C. Schulz, J. Phys D: App. Phys, 41 (2008) 055203.
[3] B. F. Kock, C. Kayan, J. Knipping, H. R. Orthner, P. Roth, Proc. Comb. Inst., 30 (2005) 1689.

LII and one-wavelength Aethalometer measurements of particulate matter in different environments

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Laser-Induced Incandescence (LII) technique is a powerful tool to measure concentration and size of soot particulate. In this work LII measurements are performed in different experimental conditions and compared with the ones derived by using a commercial aethalometer. This instrument allows to obtain the on-time concentration of optically absorbing aerosol particles by measuring the attenuation of 800 nm wavelength light through a quartz fiber filter. The filter is blackened over time with the aerosol picked up inside the instrument at controlled flows. Measurements are carried out with one second time-resolution. Absolute measurements in the ng/m^3 range are derived for the particulate concentration. As for Laser-Induced Incandescence, soot particles are sampled in a test cell, consisting of a pyrex tube. The IR beam of a Nd:YAG laser (6 Hz, 200 mJ/cm2) is properly aligned within the tube. The LII signal is detected at two wavelengths (530 nm and 700 nm) with PMT modules coupled with interference filters. A fast digital oscilloscope, triggered by the laser Q-switch pulse, is used for data acquisition and storage.

The two sets of measurements are carried out at the exhaust of a soot generator (fuelled by methane) and of a diesel engine as well as in ambient air conditions (office and laboratories). In this way, a wide range of soot load and particulate of different nature are investigated.

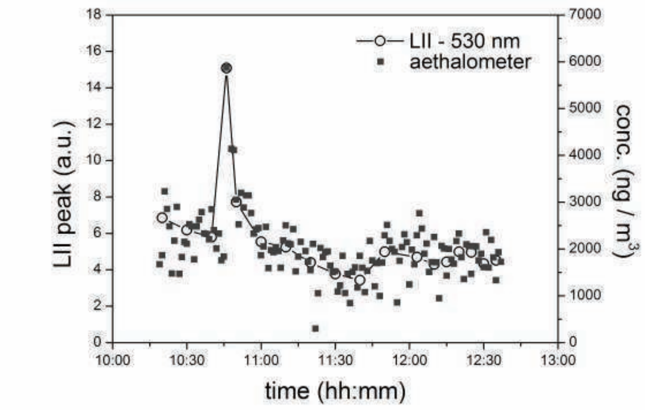

Fig. 1: *LII peak (left) and aethalometer measurements (right) versus time.*

As an example, in Fig. 1 measurements carried out in ambient air are shown versus time. Open symbols refer to the values of the LII peak at 500 nm wavelength collected about every 10 minutes. The concentration values obtained with the aethalometer are reported in closed symbols. The two sets of measurements are quite well overlapped confirming that the two techniques are sensitive to the same soot particulate and that the developed LII apparatus exhibits the high sensitivity necessary for environmental measurements.

An experimental setup to study soot sublimation as typically occurring in high fluence LII

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Particle sizing with LII requires modeling of the temporal behavior of the laser-induced emission. While most models are well validated in the low fluence regime, agreement for high fluences is identified to be insufficient, specifically when comparing different models for the short time window of soot sublimation (see Fig. 15 in [1]).

As a consequence, particle sizes are typically deduced in a low fluence time-resolved LII experiment, with the temporal fit window starting after sublimation is assumed negligible. This approach becomes inconvenient with increasing pressure when the decay rates decrease significantly towards the duration of the exciting laser pulse. Modeling of the full LII profile is then desirable requiring best possible modeling of all sub-processes involved.

Sublimation of the soot surface due to a rapid temperature increase causes a rapid vapor expansion of approximately 3-4 orders of magnitude within few nanoseconds. This correlates with the assumption of a supersonic expansion once vaporization becomes effective (see eq. 70 in [2]) and the related audible sound. To confirm this assumption, attempts can be made to detect and characterize the resulting blast wave. An example is identified in [3] where the expansion causes beam steering of a monitor laser beam passing a pulsed laser heated soot volume.

Our experiment makes use of an experimental setup used to study plasma ignition of sprays [4,5]. A green laser pulse is focused into a premixed sooting flame and the resulting effect is monitored with a Schlieren setup involving an intensified CCD camera for detection. Because the expansion of the created wave produces a very weak gradient in our current setup, we had to use very high laser fluences, clearly beyond typical LII applications. However, the expansion occurring at LII employing “high fluences” is expected to follow a similar behavior as that detected for fluences close to plasma generation in flames. The wave expansion in the flame is somewhat faster than speed of sound at the respective flame temperatures while extrapolation to the wave origin is not possible at the required accuracy.

Based on this first approach we present ideas to optimize the experimental setup for future experiments then suited to validate the assumptions currently employed in calculating the sublimation term in LII models.

[1] H.A. Michelsen et al., Appl. Phys. B **87**, 503-521 (2007).
[2] H.A. Michelsen, J. Chem. Phys. **118**, 7012-7045 (2003).
[3] K.A. Thomson et al., Proc. Combustion Institute Canadian Section 2011 Conference, Paper CICS11-39, Manitoba, Canada, 2011.
[4] G.C. Gebel et al., Proc. Fifth European Combustion Meeting, Paper 061, Cardiff, 2011.
[5] G.C. Gebel et al., Proc. ASME Turbo Expo 2012, Paper GT2012-68963, Copenhagen, Denmark, 2012.

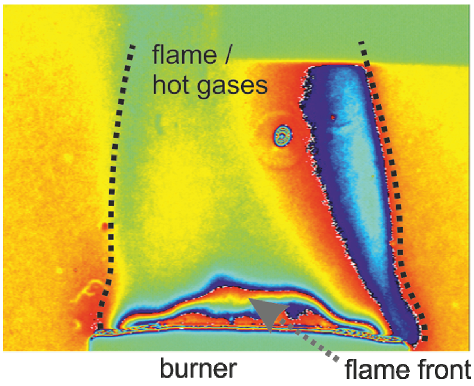

Fig. 1: Exemplary picture from time series visualizing an expansion wave generated by a high fluence laser pulse, detection delayed by 1.25 µs.

Time-Resolved Laser Induced Incandescence Measurement for a Combustion Field of the 0.5 kg-coal/h Pulverized Coal Jet Burner

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Hiroaki Watanabe[2] Hisao Makino[2] Fumiteru Akamatsu[1]

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To elucidate soot formation characteristics of pulverized coal combustion, the time-resolved laser Induced Incandescence (TIRE-LII) measurement is adapted to the 0.5 kg-coal/h pulverized coal jet burner. Whereas the TIRE-LII can be utilized to two-phase combustion phenomena, as far as we know, there have been no reports on measurements of primary soot particle size distribution in pulverized coal flames employed the TIRE-LII. The purpose of this study is, therefore, to apply the TIRE-LII measurement to a pulverized coal flame in order to elucidate soot formation characteristics in pulverized coal flames. The primary soot particle size distribution in the lab-scale pulverized coal flame, which stabilized by methane-air diffusion flame, is measured by the TIRE-LII.

Figure 1 shows a schematic illustration of the experimental set up. TIRE-LII signals are detected by using two high-speed CMOS cameras (Vision Research Inc., Phantom V5.0) located vertically to the laser sheet. The time interval between the first acquisition and the second acquisition of LII signals for TIRE-LII is set to 450 ns. In order to avoid signals' interference other than LII signal, the detection timing and the wavelength are controlled by the optical interference filter and a pulse delay generator, respectively. TIRE-LII measurements are conducted at 135 mm height from the burner tip and the ensemble-averaged values of primary soot particle size distribution are measured. The ensemble-averaged values are obtained using 500 measurements.

Figure 2 shows the instantaneous two dimensional LII signal intensity distribution (the gray-scale image) and the time-averaged primary soot particle size distribution. It can be understood from Fig. 2 that isolated soot formation areas are formed in the coal flame. In addition, the primary soot particle size distribution in the pulverized coal flame is similar to that in diesel engines.

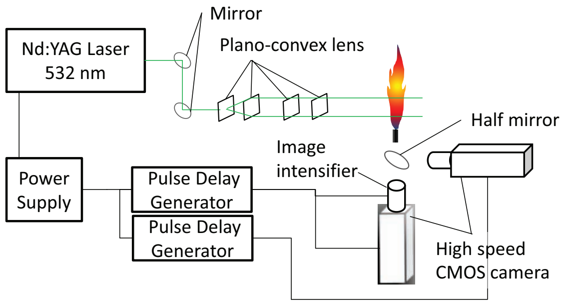

Fig.1 A schematic illustration of the experimental set up

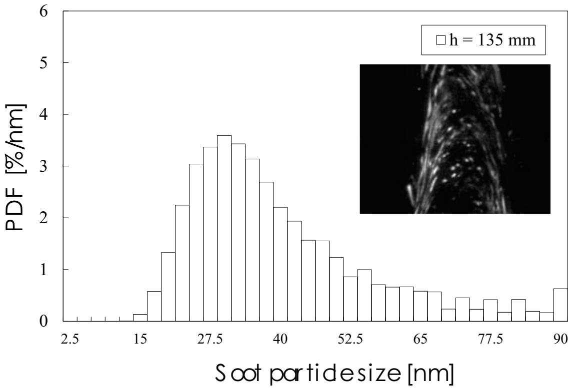

Fig.2 Soot particle size distribution and instantaneous LII signal intensity distribution of pulverized coal flame

Determination of the dimensionless extinction coefficient for soot generated by a PMMA flame

Damien Hebert [1], Alexis Coppalle [1], Jérôme Yon [1] and Martine Talbaut [1]

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Experimental soot concentration data in flames are useful for the validation of soot production and radiation models. Among the experimental methods available in order to determine soot volume fractions in fluid flows, Laser Induced Incandescence (LII) is a powerful method allowing to determine local soot volume fractions. Now, this technic is mature enough to be applied to more and more complex situations including in the field of fire studies, as in the present study. LII has been applied to the determination of soot volume fraction fields in the flame of a vertical PMMA slab. Indeed, during solid material combustion, the fuel pyrolysis is a key phenomenon, which depends on the heat transfer between the flame and the unburnt material [1]. So, as a source of radiation, soot particles play an important role in the solid material combustion.

An important step of the LII signal analysis is the calibration, in order to determine the relationship between the LII signal and the soot volume fraction [2]. The most-used approach for calibration consists in measuring the light extinction coefficient K_{ext}, which depends on soot volume fraction f_v. K_{ext} is also a function of soot particle morphology and of the optical index of the soot matter, which varies with the wavelength. But much of the optical index or K_{ext} data for soot have been determined from measurements in gaseous or liquid fueled flames but few for the solid combustion. Additionally, K_{ext} has been usually determined ex-situ for soot samples at ambient temperature [3]. Therefore one can wonder if the soot optical properties at standard or flame temperatures can be considered similar. The present work focuses on this question.

In this context, the spectral value of K_{ext} has been measured by using in-situ extinction measurements with a white laser beam (Leukos) crossing the flat flame of a PMMA slab. With the same experimental setup, K_{ext} is also determined in a gaseous fueled flame generated by a bronze porous burner and fed with a mixing of methane and ethylene. So the determined K_{ext} coefficients are relevant of the soot optical properties at high temperatures. In order to observe the influence of the soot temperature on the spectral variation of K_{ext}, it has been also determined by ex-situ measurements after sampling of soot in the flame [4]. This sampling has allowed additional measurements, the agglomerate soot particle size using a Scanning Mobility Particle Sizer (SMPS) and the mass concentration with a Tapered Element Oscillating Microbalance (TEOM).

The experimental results are analyzed to compare the spectral variations of K_{ext} in the 400-1100 nm range. Finally, different evaluations of the dimensionless extinction coefficient $K_e=K_{ext}/f_v$ are proposed corresponding to soot generated by gaseous or solid combustion at standard or flame temperatures. A quantitative comparison with the values found in the literature is presented.

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Influence of soot aggregate structure on particle sizing using laser-induced incandescence

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Soot aggregates formed in combustion processes can be described as random fractal structures. For theoretical studies of the physical properties of such aggregates, they have often been modelled as spherical primary particles in point contact. However, transmission electron microscopy (TEM) images show that the primary particles in general are more connected than in a single point; there is a certain amount of bridging between the primary particles. The results of particle sizing using laser-induced incandescence (LII) is crucially dependent on the heat conduction rate from the aggregate, which, in turn, depends on the amount of bridging.

In this work, aggregates with bridging are modelled using overlapping spheres, see Fig. 1, and it is shown how such aggregates can be built with specific fractal parameters. Aggregates with and without bridging are constructed, and it is investigated how the bridging influences the heat conduction rate in the free-molecular regime. It is shown that bridging has a significant influence on the shielding parameters that are inferred from the heat conduction results, Fig. 2. These results are used together with an LII model to show how LII particle sizing is affected by the difference in bridging.

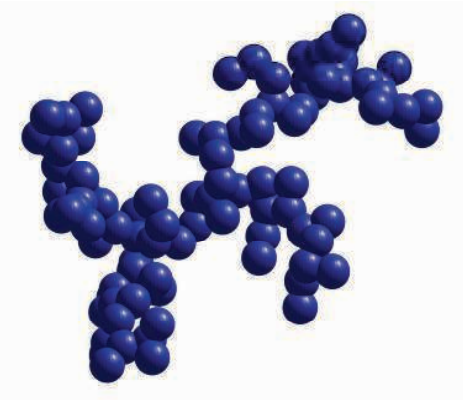

Figure 1. Example aggregate with 100 primary particles and 25 % bridging (k_f = 2.3 and D_f = 1.8).

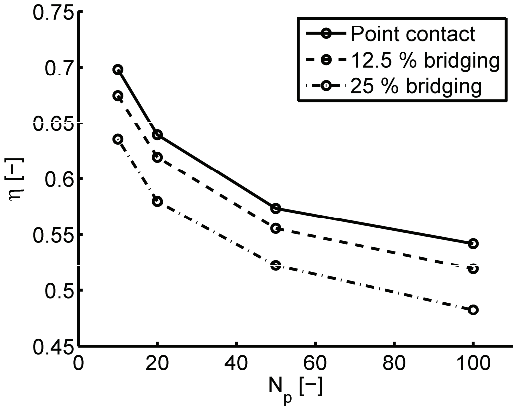

Figure 2. Example of shielding values, η, for aggregates with point contact and with bridging. N_p denotes the number of primary particles per aggregate and the heat accommodation coefficient is here set to α_T = 1.0.

Soot particles detection by LIBS and LII analysis

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Laser-Induced Breakdown spectroscopy (LIBS) is an atomic emission spectroscopy technique that has been used for elemental analysis of solid, gaseous and aerosol samples. The LIBS technique involves a pulsed laser beam focused onto the sample to create a microplasma. The resulting optical breakdown decomposes and excites all species within the plasma volume. The light emission is characterized by a continuum spectrum (Bremsstrahlung) containing discrete atomic emission lines. Both the continuum spectrum and the atomic lines decay with time. In general, the continuum spectrum decay faster than the atomic lines allowing the possibility of detecting atomic lines with a good signal-to-noise ratio by adjusting the delay and the integration time of the detector gate.

In this work the applicability of the LIBS technique to the detection of carbonaceous particulate in a combustion environment is investigated. In particular, a comparison of the carbon atom concentration derived with LIBS and LIImeasurements is performed. Soot particles produced by an ethylene-fueled soot generator are sampled and pumped for LIBS analysis into an optically equipped sample chamber, the outlet of which is then piped in the LII measuring equipment. As for LIBS the IR beam of a Nd:YAG laser was focused to create the plasma and the relative spectral emission was collected onto a fiber bundle coupled to a spectrograph-ICCD unit. As for LII, an home-made portable instrument has been used. The LII signal is detected at two wavelengths (530 nm and 700 nm). A fast digital oscilloscope is used for data acquisition and storage.

Since in LIBS technique the emission line is attributed to a particular atomic element whatever is the initial molecular species containing that element, particular care has to be taken in applying the technique to a combustion environment. In fact, in the case of carbonaceous particles, the elemental analysis does not allow to discriminate the contribution of soot particle and gas-phase species to the carbon atoms measured. The aim of the work is to develop a new methodology to select the contribution of soot particles carbon atoms in the LIBS signal.

In order to discriminate the contribution from soot particles in LIBS signals, a proper choice of the laser operating condition is performed. The laser energy is reduced to a value such that in a pure gas phase environment no breakdown is produced and the LIBS signal is induced by the presence of particles in the probe volume. However, even in these conditions carbon atoms coming from gas species are activated as well. To discriminate the two contributions to the LIBS signal a comparison is carried out between measurements below and above the breakdown threshold. The results confirm that the LIBS technique, applied with the developed procedure, is able to detect soot particles measuring soot concentration in agreement with LII measurements.

Pulsed laser heating of differently aged soot probed using LII and ELS

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Laser-induced incandescence (LII) is a laser-based technique for measuring soot particle sizes and soot volume fractions e.g. in flames, furnaces and exhaust gases. The basic principle of the technique is a rapid heating of soot particles with pulsed laser light and then detecting the increased incandescence. During the last two decades LII has been extensively refined and evolved to one of the standard techniques when conducting in situ measurements on soot particles. However, there are still refinements to be made, not least in understanding the interaction between laser light and soot, which is crucial information in the evaluation process where this interaction is modeled together with the subsequent cooling of the soot particles.

In this work the heating and vaporization effects of the LII laser pulse on differently aged soot have been studied by a combination of LII and elastic light scattering (ELS) with an experimental procedure similar to the one used in [1], but with additional possibility for pyrometry using two-color LII. By using an Nd:YAG laser at 1064 nm for the LII and another at 532 nm for ELS, and intersecting the laser beams in the probe volume, the elastic scattering can be utilized to probe heating and vaporization effects of the 1064 nm laser light. To generate the soot particles a McKenna type burner has been used, where the soot growth can be followed from nascent soot particles close to the burner surface to more mature soot higher up in the flame.

Results show a significant difference as a function of height above burner (HAB) as seen in Fig. 1. This behavior could be explained by a variation in both the absorption function, E(m), and the sublimation threshold at different HAB i.e. for differently aged soot. Furthermore the results reveal an effect not discussed in [1], namely a slight increase of the scattering signal from soot just before the rapid decrease due to vaporization, seen in Fig. 2.

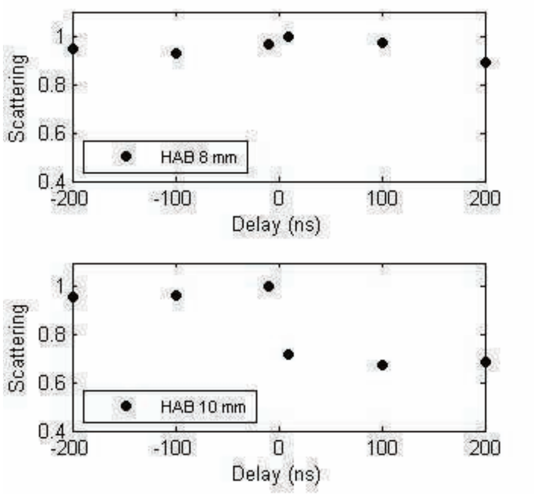

Figure 1 Normalized ELS signal before and after an LII laser pulse with fluence 0.3 J/cm^2. Top plot shows measurement at 8 mm HAB and bottom plot at 10 mm HAB.

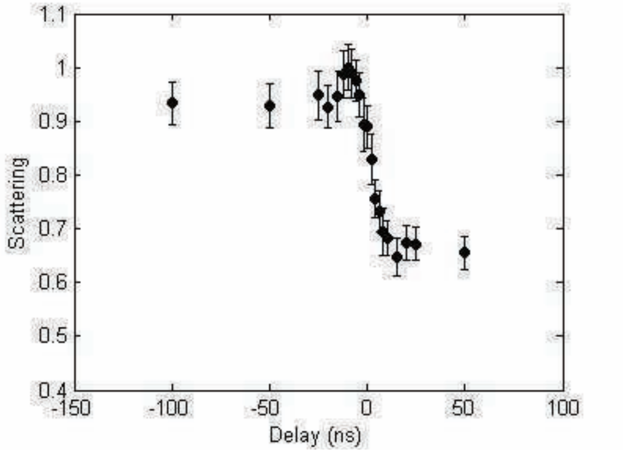

Figure 2 Normalized ELS signal before and after an LII laser pulse with fluence 0.3 J/cm^2 at 10 mm HAB.

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Recent applications of the WALS-technique

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Nanoparticles produced in combustion processes often exhibit complex fractal structures. While laser-induced incandescence (LII) is a proven technique for the determination of primary particle size no information about aggregate sizes can be obtained. To gather information about aggregate size and fractal dimension elastic light scattering (ELS) [1] is an often used *in situ* method.

The wide-angle light scattering (WALS) approach [2] extends classical ELS-concepts by using a combination of an ellipsoidal mirror and an intensified CCD-camera. The ellipsoidal mirror redirects the light scattered within a plane onto the CCD-chip (cf. Fig. 1), which makes it possible to almost instantaneously record a complete scattering diagram over an angular range of approx. 10° to 170° with an angular resolution $\Delta\theta$ of typically 0.6°.

The basic performance of the approach was demonstrated previously by measurements on soot particles in laminar premixed flames [2]. This contribution highlights various recent developments and applications of the technique. These include measurements in a turbulent diffusion flame [3], employing a pulsed laser and underlining the favourable applicability to unsteady processes. Also measurements with a particular high resolution of $\Delta\theta = 0.3°$ were performed which allow for a detailed investigation of selected angular regions. To simultaneously measure the vv- and hh-scattering components polarization foils were mounted in front of the ellipsoidal mirror. Radii of gyration obtained for soot particles in a premixed ethene flame show good agreement with former results. Furthermore investigations on silica particles produced in a diffusion flame were carried through (cf. Fig. 2) for various relative velocities between the precursor flow (nitrogen flow saturated with hexamethydisiloxane) and the methane/oxygen flow of the supporting flame. Recorded scattering diagrams indicate a change in the structure of the silica particles for the different velocities.

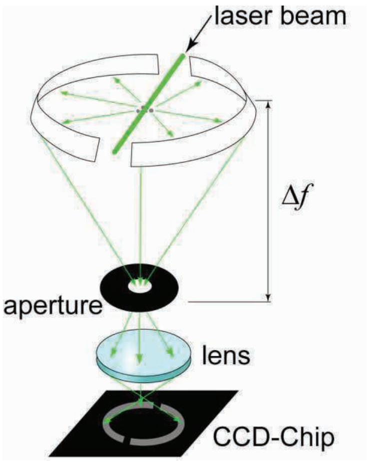

Fig. 1: *Experimental setup*

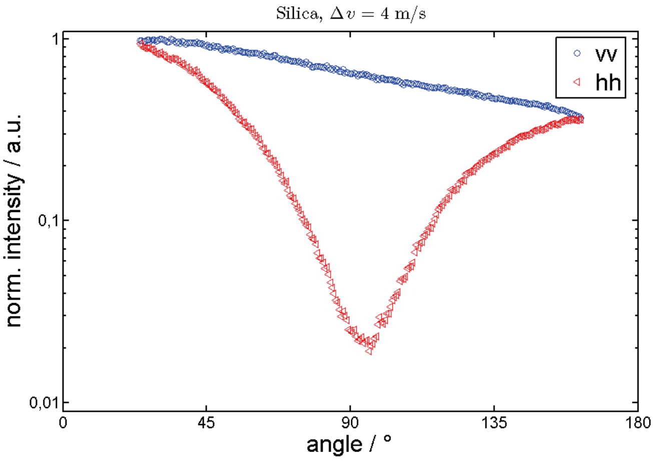

Fig. 2: *Measurement on silica particles in a diffusion flame*

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A combined laser induced incandescence, aerosol mass spectrometry, and scanning mobility particle sizing study of non-premixed ethylene flames

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Investigations into the chemical composition of soot particles often rely on samples extracted from the flame environment for subsequent analysis by mass spectrometry. These mass spectra have contributed to the general consensus that polycyclic aromatic hydrocarbons (PAHs) are involved in soot nucleation and surface growth processes. In the pioneering work of Dobbins et al.,[1,2] soot and soot precursor particles were extracted along the centerline of a non-premixed coflow flame by rapid-insertion thermophoretic sampling and subsequently analyzed by laser microprobe mass spectrometry (LMMS). Low in the flame, where TEM images suggested the particles were liquid-like, the mass spectrum was dominated by species between 200 and 300 amu in size. Higher in the flame, where TEM images indicated that carbonaceous aggregates were being formed, PAHs were no longer observed in the mass spectra. The laser-desorption ionization experiments of Bouvier et al.,[3] Lemaire et al.,[4] and Faccinetto et al.[5] complemented and expanded the findings of Dobbins et al.[1,2] Lemaire et al.[4] showed that the fuel composition strongly influences the soot composition. Faccenetto et al.[5] developed a new sampling method enabling some distinction between PAHs in the gas phase and those adsorbed onto the soot particles. Intrusive sampling techniques such as those used in the studies referenced above perturb the flame. In some instances such techniques may permit the agglomeration of existing particulates, condensation of low vapor pressure species onto the surface of soot nuclei, and nucleation of new clusters that could later be erroneously associated with nascent soot. Recently, we have coupled a non-premixed, opposed-flow flame system to an aerosol mass spectrometer to investigate the chemical composition of soot particles extracted from different regions of the flame. The present work provides insight into the effects of our intrusive sampling method on the observed soot composition and size distributions by combining laser-induced incandescence (LII) measurements with intrusive particle diagnostics.

In this work, we performed time-resolved LII measurements in conjunction with flame-sampling aerosol mass spectrometry (AMS) and scanning mobility particle sizing (SMPS) measurements to (1) investigate differences in particle size and composition as a function of temperature and position in the flame and (2) investigate processes occurring within the sampling system during sample extraction. We probed three non-premixed, opposed-flow, ethylene flames at conditions ranging from nearly sooting to moderately sooting. In situ time-resolved LII measurements revealed differences in the temporal response as a function of laser fluence and position in the flame. These differences may result from varying absorption coefficients due to particle composition and/or differences in the extent of particle surface coatings. Particles extracted from different regions of the flame also yielded varying temporal LII profiles, size distributions (as determined by SMPS), and chemical compositions (as determined by AMS). The use of a thermal denuder prior to the ex situ LII, SMPS, and AMS instruments also provided information on the extent of PAH condensation onto existing particles and the propensity for liquid- or tar-like droplet nucleation within the sampling system.

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A method for inferring the soot size distribution by Static Light Scattering : Application to the CAST soot generator

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The soot size distribution is often determined by using ex-situ granulometers, after sampling of the particles. But the quenching of aggregation process during the sampling is difficult to control and raises the question of representativeness of the results (Ouf et al. 2010 [1]). For this reason, optical measurements are more suitable. Thanks to the approximation of Rayleigh – Debye – Gans for Fractal Aggregates which is well suitable for such particles, (RDG-FA theory - Dobbins et al., 1991 [2]), measurement of static light scattering (SLS) can be interpreted in order to determine a size parameter called gyration radius. The inversion of experimental data by this theory to infer the gyration radius of monodisperse aggregates has been recently validated (Caumont et al. 2010 [3]).

The SLS technique consists in measuring the signal scattered by the particles after interaction with a polarized laser light (532 nm in our case) at different scattering angles. Since a long time, some authors have proposed to determine a representative gyration radius of the polydisperse population with this optical technique (Dobbins & Megaridis 1991 [2], Köylü 1994 [4]). More recently, inversion methods have been proposed by coupling scattering and extinction measurements (Koylu & Faeth (1996) [4] and Iyer et al. (2007) [5]). But these methods rely on the knowledge of soot optical index which is unfortunately not accurately known. Moreover Burr et al [6] by using Bayes' theorem showed mathematically that the inverse problem is ill-posed..

This work presents a new inversion method for the determination of soot size distribution in flames by measuring scattered light at different angles. It consists in determining for each studied angle, by using the RDG-FA theory, a gyration radius $R_g^*(\theta)$ of a monodisperse population which has the same optical behavior as the real polydisperse population. The so determined $R_g^*(\theta)$ function informs us about polydispersity of soots. The method has been validated by comparing obtained results with size distribution determined by Transmission Electron Microscopy (TEM). The method is now applied to characterize soot size distribution generated by the Jing CAST apparatus that is suspected to become a standard for soot generation including for LII calibration. Results are compared with soot size distribution determined by DMS500 apparatus.

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List of posters

(alphabetic order first author)

Sizing of soot aggregates by two-dimensional multi-angle light scattering (2D-MALS)
Michael Altenhoff, Jannis Reisky, Stefan Will
Universität Erlangen-Nürnberg, Germany Universität Bremen

Study of the wavelength dependence of the soot absorption function using the two-excitation wavelength Laser Induced Incandescence: application to fluorescent species detection.
S. Bejaoui, R. Lemaire, E. Therssen, P. Desgroux
PC2A, University of Lille 1, Ecole des Mines de Douai

Comparison between Modeled and Measured Time-Resolved LII Signals and Soot Temperatures in a Laminar Premixed Flame
S. Bejaoui, S. Batut, E. Therssen, P. Desgroux, F. Liu, K. A. Thomson, G. J. Smallwood
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John D. Black and Paul Wright
University of Manchester

Applicability of Wright's correction to Fuchs boundary sphere method for TiRe-LII calculations
K.J. Daun, S.C. Huberman
University of Waterloo

Influence of temporal laser pulse length and shape on the time resolved laser induced incandescence signal
M. Ditaranto, N.E. Haugen, C. Meraner, I. Saanum
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Combined LII and LIF with multiple excitation wavelengths for diagnostics of soot and PAHs in laminar flames
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Experimental study of particle vaporization under pulse laser heating by LII and laser light extinction
Alexander Eremin, Evgeny Gurentsov, Ekaterina Mikheyeva, Konstantin Priemchenko
Joint Institute for High Temperature, Russian Academy of Sciences

Experimental investigation of the influence of inert gas on soot formation
A.Flügel, S. Beer, S. Will, J. Kiefer, A. Leipertz
University of Erlangen-Nürnberg, SAOT Erlangen, University of Aberdeen

Combination of high spatial resolution LII and LOSA measurements for determination of soot volume fraction and PAH concentration in laminar diffusion flames
M. Leschowski, K. Thomson, D. Clavel, D. Snelling, C. Schulz, G. Smallwood
IVG and CENIDE, University Duisburg-Essen, NRC Ottawa

Combination of various particles measurement techniques for validation in laminar high pressure flames
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IVG and CENIDE, University Duisburg-Essen

Effect of laser pulse duration on laser-induced incandescence soot
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NRC Ottawa

Measurements of Soot Volume Fraction under Conditions Relevant to Engine Exhaust Using Four-Colour LII and Different Laser Energies
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NRC Ottawa, Beijing Institute of Technology

Relationship between LII Signal and Soot Volume Fraction – Effect of Primary Particle Diameter Polydispersity
Fengshan Liu, Gregory J. Smallwood
NRC Ottawa

Novel soot volume fraction measurement through ratio-pyrometry and absolute light calibration
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Yale University

Modeling laser-induced incandescence of soot integrating spatial and temporal dependences of parameters involved in energy and mass balances
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Ecole des Mines de Douai

Evolution of the LII signals of soot particles measured in low pressure methane flames
T. Mouton, P. Desgroux, X. Mercier
PC2A, University of Lille1

Influence of LII on Soot Optical Properties in Reference Flames
K. Thomson, K-.P. Geigle, D. Snelling, F.Liu, M. Köhler, G. Smallwood
NRC Ottawa, DLR Stuttgart

Evaluation of particle sizes of iron-oxide nano-particles in a low-pressure flame-synthesis reactor by simultaneous application of TiRe-LII and PMS
B. Tribalet, A. Faccinetto, T. Dreier, C. Schulz
IVG and CENIDE, University Duisburg-Essen

Comparison of different techniques for measurement of soot and PM emission from Diesel engine
Richard Viskup
Institute for Design and Control of Mechatronical Systems,Johannes Kepler University

Sizing of soot aggregates by two-dimensional multi-angle light scattering (2D-MALS)

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For the understanding of soot formation in combustion processes comprehensive information about local size properties of complex soot aggregates is desired. Elastic light scattering (ELS) is a well-established optical technique which allows for the *in situ* determination of aggregate size and fractal dimension of soot particles in flames [1]. Reimann et al. [2] used a two-dimensional combination of ELS and laser-induced incandescence (LII) for the determination of various parameters of soot particles in a premixed flame from a porous flat flame burner (McKenna type). Although the general approach was successful both the measuring range in terms of aggregate size and the information obtained were limited because of the use of a fixed scattering angle of 90°.

In continuation and extension of this approach we performed two-dimensional ELS-measurements under various scattering angles thus allowing for a simultaneous acquisition of particle parameters at various heights above burner (HAB). Measurements on a premixed ethene flame from a McKenna type burner with an equivalence ratio of 2.7 were carried out by irradiating a laser-light-section and detecting the scattered light using an intensified CCD camera (cf. Fig 1). The detection angle varied equidistantly in the scattering vector q from 17° to 163°, and the evaluation of obtained data was carried out for each pixel line from 10 mm to 20 mm HAB for three different areas: the flame axis only, the area determined by the depth of field and the maximum evaluable region. The obtained radii of gyration show good agreement with former results.

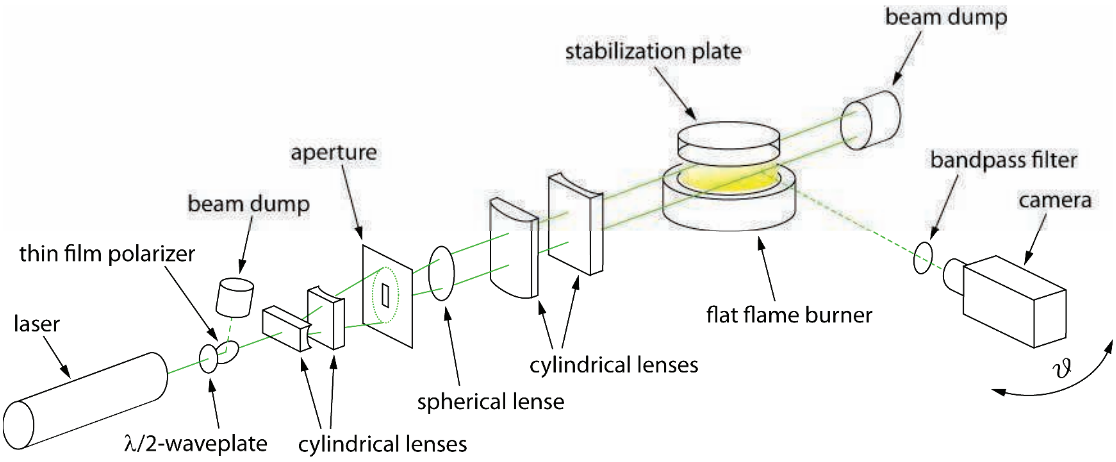

Fig. 1: *Experimental setup*

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Study of the wavelength dependence of the soot absorption function using the two-excitation wavelength Laser Induced Incandescence: application to fluorescent species detection.

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In this work, wavelength dependence of the absorption function of soot was experimentally studied. We used a technique developed in our team which consists to heat similarly the soot particles using two different laser excitation wavelengths. Thus, using two lasers with the same temporal and spatial irradiance profiles, it is possible to find combinations of the both lasers energies, below sublimation regime activation, insuring that soot particles absorb the same energies, reach the same temperature and emit the same Laser Induced Incandescence (LII) radiation [1]. Laser at 1064 nm is always chosen as a reference excitation and compared with a UV-visible wavelength (λ_i) such as 266 nm, 355 nm, 532 nm. In this way we can deduce the relative evolution of the absorption function $\frac{E(m,\lambda_i)}{E(m,1064nm)}$ versus wavelength.

Experiments are investigated in a turbulent diffusion flame of pulverised diesel and in a premixed methane flame stabilized on a McKenna burner. It is found that up to 700 nm the emission signal due to PAH /soot precursor LIF interferes with the LII one. Interestingly no LIF emission could be identified above 700 nm. Therefore this spectral region appears very attractive to collect soot incandescence in flames containing PAH.

The two-excitation wavelength LII method has been checked for the first time using a narrow spectral detection set above 700 nm, by using several combinations of UV-visible and IR radiations. Soot particles heating was controlled either looking at the Planck function above 700 nm or controlling the decay rate of the LII temporal signals. Once similar heating is reached using any UV-vis radiation and the 1064 nm one, the method is used to get either the ratio of soot absorption functions, or the LIF spectra of soot precursors even in the presence of soot.

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Comparison between Modeled and Measured Time-Resolved LII Signals and Soot Temperatures in a Laminar Premixed Flame

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LII experiments were conducted in a laminar premixed flame established with a McKenna burner at atmospheric pressure burning a mixture of methane, oxygen, and nitrogen with an equivalence ratio of 2.15 and with a flame stabilizer of stainless steel plate placed at 20 mm above the burner. A Nd:YAG laser at 1064 nm with a repetition rate of 10 Hz was used to produce a top-hat laser beam with a 6 ns pulse duration (FWHM) as the excitation source. Time-resolved LII signals were measured at 610 nm (20 nm FWHM) using a PMT at different locations in the flame and at different laser fluences. Soot temperature measurements were also conducted through recording LII spectra with a spectrograph at different locations along the flame centerline and different laser fluences. TEM analyses of soot sampled at HAB = 12 and 15 mm were also carried out to provide primary particle diameter distribution and average number of primary particles in an aggregate for LII model calculations.

Preliminary model calculations suggest that the base model LII developed at NRC was able to reproduce the experimental resolved LII signals accurately in the low-fluence regime; however, large discrepancies between the model and the experimental results occur at high fluences. To understand the role of physical and chemical processes that were not incorporated into the NRC LII model, annealing and photodesorption were implemented and their effects on the LII model results were investigated. This study is aimed at improving the NRC LII model at high laser fluences through a detailed comparison between the experimental LII results and the modeled LII results.

Continuous Wave LII in an Atmospheric Pressure Kerosene Flame

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Fibre and diode lasers with sufficient power to heat soot particles to incandescent temperatures are readily available at lower cost than the nanosecond pulsed lasers traditionally used in LII. There are less stringent safety restrictions on the use of CW lasers and they can be delivered with excellent beam quality through standard optical fibres, making them more suitable for LII in practical environments. Using the collimated beam from a diode laser at 803 nm in the power range 5 – 30 W, LII was easily observable in a highly sooting kerosene flame (F_v ~10^{-5}). However, the laser causes major changes in the combustion, increasing soot burn out rates and transferring heat to other regions of the flame.

In contrast to short pulse LII, soot particles experience laser heating and cooling by heat transfer at rates comparable with their reaction rate. Their residence time in the beam and other processes such as photophoresis and optical trapping also have to be considered. Hence, modeling is much more complicated than for short pulse LII, and the processes are not well understood.

Figure 1: LII in a quasi-2-D kerosene lamp flame with 28 W 1 mm diameter cw laser beam photographed through a BG3 filter

Visible emission spectra were collected using a traversable fibre optic probe from a magnified projected image of the flame shown in Figure 1. There is a good match between predicted emission spectra based on the blackbody curve and observed spectra from the flame in the wavelength range 590 – 790 nm. From these spectra estimated soot temperature in the absence of the laser is 2150 K, rising to 2600 K in the region of a 28.5 W laser beam. Temperature rise is linear in laser power. Local soot temperature is increased both above and below the beam when the laser is present. Above the laser beam, light emitted at 700 nm decreases quadratically with distance from the height of the centre of the laser beam to the edge of the visible flame, although the soot particle surface temperature remains at ~2350 K in this upper part of the flame. The intensity of light emitted at 700 nm at the centre of the laser beam at varying laser power is in good agreement with a prediction based on blackbody radiation. This indicates that the mechanism of increased light emission is particle heating (LII) and not creation of additional soot by laser stimulated reactions in the flame.

Although cw LII is at a very early stage of development, the potential for combustion diagnostics – soot concentration, temperature, velocity by flow tagging, etc. – is obvious. The observations described here should provide a basis for future investigation of the processes involved.

Applicability of Wright's Correction to Fuchs' Boundary Sphere Method for TiRe-LII Calculations

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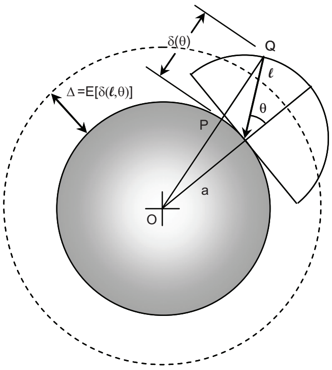
When LII experiments are performed on high pressure aerosols, transition-regime heat conduction from the laser-energized particles is usually calculated using Fuchs' boundary sphere method. In this technique the Knudsen layer is represented by a collisionless boundary sphere enveloping the particle, which in turn is surrounded by a continuum gas. The analysis proceeds by equating the heat transfer through the two domains and then solving for the unknown boundary sphere temperature, T_Δ.



This calculation requires specification of the spherical shell thickness, Δ, which is usually chosen as the mean free path at T_Δ, $\lambda_\Delta = \lambda(T_\Delta)$. Filippov and Rosner [1] instead advocate a more complex equation that accounts for particle curvature and the directional distribution of incident molecules, originally proposed by Fuchs [2] and derived by Wright [3] to model evaporating droplets. If a colliding molecule has travelled a distance ℓ from its most recent collision at an angle θ relative to the surface normal, the corresponding radial distance is $\delta(\ell,\theta) = (\ell^2+a^2+2\ell a\cos\theta)^{1/2} - a$. By integrating over all incident angles, the expected value of $\delta(\ell, \theta)$ for a given ℓ is

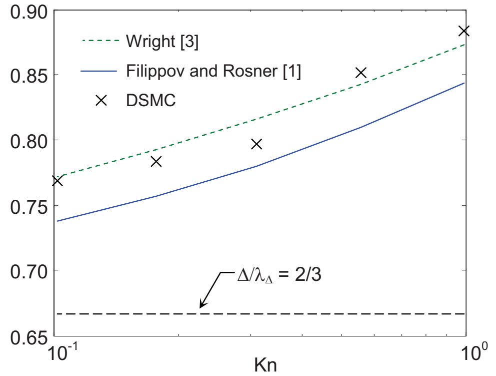
$$\delta(\ell) = \int_0^{\pi/2} \delta(\ell,\theta) P_\theta(\theta)\, d\theta = \frac{a^3}{\ell^2}\left[\frac{(1+\ell/a)^5}{5} - \frac{(1+\ell^2/a^2)(1+\ell^2/a)^3}{3} + \frac{2}{15}(1+\ell^2/a^2)^{5/2} - \frac{\ell^2}{a^2}\right] \quad (1)$$

where $P_\theta(\theta) = 2\cos\theta\sin\theta$. Filippov and Rosner [1] set $\ell = \lambda_\Delta$ in Eq. (1) to find Δ, while Wright [2] also considers the distribution of incident paths, $P_\ell(\ell) = 1/\lambda_\Delta \exp(-\ell/\lambda_\Delta)$,

$$\Delta = \int_0^\infty \delta(\ell) P_\ell(\ell)\, d\ell = \int_0^\infty \delta(\ell) \frac{1}{\lambda_\Delta} \exp(-\ell/\lambda_\Delta)\, d\ell \quad (2)$$

which can be solved numerically.

We use Direct Simulation Monte Carlo to investigate this phenomenon under typical LII conditions. The Knudsen layer thickness is found by sampling the radial distance that incident gas molecules travel before they collide with the surface. The DSMC results reveal that particle curvature increases the Knudsen layer thickness compared to a flat surface ($\Delta/\lambda_\Delta = 2/3$), an effect captured by both Wright's equation [3] and Filippov and Rosner's [1] approximation. This correction has a negligible influence on transition regime heat transfer rates, however, especially considering other uncertainties involved in the calculation, so it can be safely excluded when analysing TiRe-LII data.



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Influence of temporal laser pulse length and shape on the time resolved Laser Induced Incandescence signal

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Q-switched Nd:YAG lasers are typical choice for LII, with excitation pulse length of 7-10 ns. The question of longer pulse duration was raised in Schulz, Kock et al. (2006) and there is interest in using CW laser for LII (Black, 2010). The preset study is an experimental investigation on the influence of pulse lengths, in the range 50 - 1000 ns, and temporal shapes. The measurements were made 25 mm above a laminar non-premixed ethylene flame, using a Nd:YAG laser with temporal shaping capabilities (Agilite, Continuum). The time resolved LII measurements were made by re-constructing averaged, sequentially delayed, gated (10 or 20 ns) ICCD images in radial profile or spectral modes. Except in spectral mode, the LII signal is recorded with a 10 ns bandpass filter centered at 488 nm and a 532 nm centered notch filter. By using the shaping capabilities of the laser the effect of pulse length has been varied by keeping constant either the pulse fluence or pulse energy.

The main feature that has been observed is shown in figure 1. At the start of the pulse, the LII signal builds up equally for all cases, as a result of particles absorbing energy and heating up. The LII signal for the 50 ns pulse is as expected, decaying at a rate dependent on the primary particle diameter, but when the pulse length is increased, one observes a shouldering of the signal after 50 ns (green curve). This observation is unexpected as for constant pulse length, increasing the fluence, is known to increase the decay rate (i.e. to narrow the LII signal), because of the increase in vaporization rate (Michelsen, Witze et al.). It therefore indicates that the effect of laser fluence expressed in J/cm^2 has a time scale dependency on the LII processes. A further increase in pulse length to 200 ns (red curve) shows not only a further delay in the decay, but also a rebound of the LII signal. The cause of this phenomenon is unclear, as none of the processes known to be involved in the LII theory predicts such an effect. One shortcoming of our set up is that the beam is formed into a sheet, however the spatial resolution is clearly defined by the collection optics and this phenomenon would happen at all pulse lengths if it was due to the interferences from out of focus signals. Figure 2 shows that the signal rebound phenomenon as pulse length increases is also observed at constant fluence, starting for pulses longer than ca. 75 ns. The behaviour was also detected with both a top hat and a quasi-gaussian pulse temporal profile. The spectrally and time resolved LII traces shown in figure 3 seem to exclude the effect of interferences from molecular excitation processes such as late fluorescence, and confirm the efficiency of the filtering scheme chosen in the measurements shown in fig. 1 and 2.

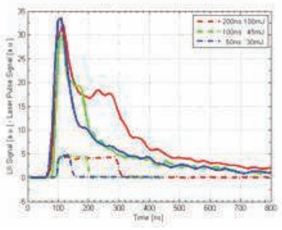

Fig. 1. Red: 200ns/100mJ; Green: 100ns/45mJ; Blue: 50ns/30mJ.

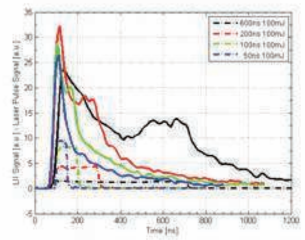

Fig. 2. E: 100mJ. Black: 600ns; Red: 200ns; Green: 100ns; Blue: 50ns.

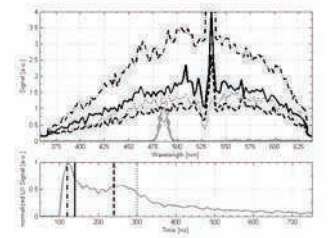

Fig. 3. Time resolved spectra taken at LII time shown in graph below.

Combined LII and LIF with multiple excitation wavelengths for diagnostics of soot and PAH in laminar flames

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Through the use of two complimentary laser techniques, laser induced incandescence and laser induced fluorescence, the formation of soot from polycyclic aromatic hydrocarbons (PAH) has been studied. Information on soot volume fraction (from signal peak intensity) and particle cooling rate (from the signal decay time) is obtained from incandescence signal, while fluorescence measurements offer information on PAH present. These techniques were used to study a premixed flat flame of ethylene and air at a range of equivalence ratios. This project involves the use of three different excitation wavelengths (1064 nm, 532 nm, 290 nm). The fluences were adjusted so that the soot particles are heated to the same temperature by each excitation wavelength, resulting in equal incandescence intensity. Since no fluorescence is detected for 1064 nm excitation the contributions from incandescence and fluorescence can be separated. This is achieved by subtracting any signal obtained for 1064 nm excitation from the signals obtained for excitation at shorter wavelengths, leaving the remaining signal to be attributed to fluorescence. A monochromator has been used to resolve the signals, thus generating a time-sequence of emission spectra. This approach is helpful in identifying the contributions of LII and LIF to the signals detected.

Experimental study of particle vaporization under pulse laser heating by LII and laser light extinction

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Particle vaporization is one of the uncertainties in LII measurements of particle size or volume fraction. The common recommended threshold of energy fluence for soot vaporization is in the range of 0.1-0.2 J/cm^2. However, for non-soot particles or for carbonaceous particles different from soot, the vaporization threshold might be unknown. One of the reasons is the decreasing of the particle refractive index function *E(m)* with particle size [1]. Due to this effect the particle heat up temperature couldn't reach the vaporization threshold at low fluences. Besides that, the vaporization temperature could decrease with particle size. The knowledge about particle vaporization process is useful not only for correct LII measurements, but also for the determination of particle thermodynamics properties changing due to size effect [2]. Direct characterization of vaporization process of soot was performed by simultaneous scattering and LII registration [3], by emission spectroscopy [4] and by two pulse lasers [5] in flames. The goal of this work is the application of laser light extinction measurements and Ti-Re LII for observation and analysis of vaporization process of small carbon and iron particles.

The carbon particles were formed in pyrolisis of 1% C_6H_6+Ar behind reflected shock wave. Two-color Ti-Re LII technique was applied for particle heat up temperature and size measurements at fluencies around 0.4 J/cm^2. He-Ne laser beam was adjusted coaxially to the YAG (1064 nm) laser beam and allowed to observe the decreasing of a volume of condensed phase due to vaporization. Additionally, the real particle temperature equilibrated with bath gas during pyrolisis process was measured by emission-absorption spectroscopy in visible range of spectra. The measured gas-particle temperature was less than frozen temperature behind shock wave due to endothermic effect of C_6H_6 decomposition. The vaporization temperature of small growing carbon particles with mean diameters of 2-14 nm was found to be in the range of 2900-3100 K in contrast to soot vaporization temperature 4000 K [6].

Growing iron particles of different sizes (2-11 nm), synthesized in the laser photolysis reactor [1], were heated by YAG laser pulse with fluences of 0.025-0.7 J/cm^2. The same technique as for carbon particles was used for condensed phase loss, temperature and size measurements. The essential difference of iron particles vaporization temperature (2100-2700 K) from the bulk one (about 3050 K) in dependence on particle size and pressure of a bath gas was found.

The dispersion of vaporization temperature observed in both experiments is probably caused by with the variation of particle properties formed at different conditions. The related value of evaporated fraction of condensed phase and particle vaporization temperature are analyzed in dependence on experimental conditions.

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Experimental Investigation of the Influence of Inert Gas on Soot Formation

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The formation of soot in non-premixed flames is influenced by many parameters, and the detailed mechanisms are yet to be fully understood. Adding inert gas to the fuel supply of a burner has an inhibitory effect on soot formation. This reduction in soot inception is probably caused by the reduction of the flame temperature and hindered air entrainment due to a modified diffusion.

In this work, laser-induced incandescence (LII) was employed in laminar propane diffusion flames at atmospheric pressure in order to study the influence of adding inert gas on soot concentration and primary particle size. A co-axial burner stabilized with an air co-flow was used to produce a stable propane flame with constant propane mass flow. The burner has a inner diameter of 13 mm, the inner diameter of the co-flow was 89 mm, resulting in Re_{fuel}=52 (for undiluted conditions) and Re_{air} = 102, respectively. The fuel was diluted with nitrogen, carbon dioxide and argon. The mass flow rate of propane was kept constant at 4.3 mg/s and the inert gas was varied from 0 to 5 mg/s in steps of 0.5 mg/s at 1 bar and 293 K. The fuel and inert gas were premixed in a T-mixer. The mixture reached the combustion zone with a laminar flow and homogeneous mixture. Laser-induced incandescence was generated by a frequency-doubled pulsed Nd:YAG laser and detected with a photomultiplier tube to obtain time-resolved LII signals. Adding 0.5 mg/s of Ar reduced the soot volume concentration to 93% of the original value, in the cases of N_2 and CO_2 the reduced values amounted to approximately 81%. Additional increments of inert gas in further steps of 0.5 mg/s resulted in the same tendency .

In consideration of primary particle sizes, nitrogen addition resulted in a considerable initial effect, while particle size in the case of carbon dioxide only changed significantly on the addition of larger flow rates.

Combination of various particle measurement techniques for validation of TiRe-LII in laminar high pressure flames

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The method of laser-induced incandescence (LII) has become a common way for in-situ particle-size measurements and visualization of particle volume fractions in a wide range of applications [1]. For the further development of LII diagnostics for high-pressure combustion systems, measurements in well-defined laminar and stationary flames enclosed in optically accessible pressure vessels [2] are necessary. Challenges for the LII technique are the quantitative determination of soot particle size and volume fraction from time-resolved (TiRe) LII signal traces under high pressure conditions and their validation by independent calibration and sampling techniques, respectively.

For a complete characterization of the flame environment, a combination of in-situ optical as well as intrusive measurements was employed in a high-pressure burner. Two-color TiRe-LII was used to determine soot particle size, which simultaneously provides the soot volume fraction from the peak intensity of the incandescence signal trace. In our experiments soot particles were heated by the fundamental of a Nd:YAG laser and the incandescence radiation was detected in two wavelength ranges by fast photomultipliers. Additionally, for the determination of soot volume fraction, laser extinction was measured in the near infrared (785 nm) to minimize interference by absorption of polycyclic aromatic hydrocarbons.

To determine the gas temperature, soot luminescence was measured spectrally resolved and fitted with a Planck function. For the spectral radiation measurement an EM-CCD camera was coupled to a spectrometer (focal length 150 mm). To independently verify particle size with transmission electron microscopy (TEM), a pneumatically-driven thermophoretic probe sampling system was designed that allows to insert TEM grids rapidly into the flame at operating pressures up to 40 bar.

The combined information contributes to further improve LII models for evaluating particle size distributions and concentration from TiRe-LII measurements.

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Combination of High Spatial Resolution LII and LOSA measurements for determination of soot volume fraction and PAH concentration in laminar diffusion flames

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Line-of-sight-attenuation (LOSA) is a well-established soot diagnostic valuable for soot volume fraction [1] and the method of laser-induced incandescence (LII) has become a common way for in-situ particle-size measurements and visualization of particle volume fractions in a wide range of applications [2].

In this work a co-annular burner was used to produce a laminar ethylene/air non-premixed flame at the standard Gülder conditions. For the LOSA measurements, a tungsten strip lamp (450–1000 nm) was used as light source. The transmission and emission signal of the flame were both detected with a spectrometer (Jarell-Ash Monospec 18), configured to measure in the wavelength range of 450–900 nm and a CCD camera (Princeton Instrument-Spectrum MM System) with a spectral resolution of 6.7 nm per pixel on the detector plane.

For the high spatial resolution LII (HSRLII) measurements a frequency-doubled Nd:YAG laser (Big Sky Ultra CFR) was used as light source. The incandescence signal was detected at 445 and 750 nm with fast photomultipliers. The probe volume had a height of 200 µm and a width of 100 µm.

The HSRLII measurements were performed as radial scans at a number of HABs. The soot volume fractions determined with HSRLII were compared to those determined with LOSA at 3 different wavelengths. For the LOSA data, the attenuation is due to soot absorption, soot precursors (PAH) absorption, and scattering. The latter two phenomena are also wavelength-dependent. As a result, the SVFs determined from the LOSA data are always slightly higher than those for HSRLII data.

With the HSRLII the volume fraction of the soot was measured precisely and with the comparison to the scattering-corrected LOSA data, the difference between the SVFs of both measurement techniques indicates the contribution of polycyclic aromatic hydrocarbons (PAHs) as soot precursors in the volume. As the HSRLII and LOSA produced radially resolved measurements at different HABs, two dimensional distributions for the volume fractions of soot and relative PAHs can be constructed. With this information the region of soot formation can also be identified. The HSRLII measurements also give the primary particle diameters for the soot particles.

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Measurements of Soot Volume Fraction under Conditions Relevant to Engine Exhaust Using Four-Colour LII and Different Laser Energies

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Available experimental evidence shows that the apparent soot volume fraction (SVF) determined from two-colour LII (at 450 and 780 nm) for soot initially at near ambient room temperature displays a laser fluence dependence: it first increases with increasing the laser fluence until it reaches a maximum, after which it decreases with further increasing in laser fluence, due to sublimation. It is suggested that the low-fluence SVF anomaly is attributed to changes in soot emissivity in the 350 to 500 nm spectral range as a result of evaporation of condensed volatile organic compounds from soot particle surfaces due to laser heating. However, there is currently a lack of direct evidence to confirm the cause of the low fluence anomaly and a lack of detection strategy on how to avoid the low fluence SVF anomaly. The current approach using the two-colour LII technique is to operate the laser fluence around 2.1 mJ/mm^2 with a 1064 nm laser, see Fig. 1.

In an attempt to address these two questions experimental measurements of SVF were conducted in a soot aerosol at near room temperatures at different laser fluence. A Nd:YAG laser of 6 ns FWHM operated at 532 nm was used to excite the soot particles. The resultant LII signals were detected at four spectral bands (with spectral widths varying between 12 to 40 nm) centered at 400, 631, 780, and 840 nm. The experimental results from this work provide useful insights into the cause of the low fluence SVF anomaly and an effective strategy to avoid this anomaly through detection of LII signals at longer wavelengths.

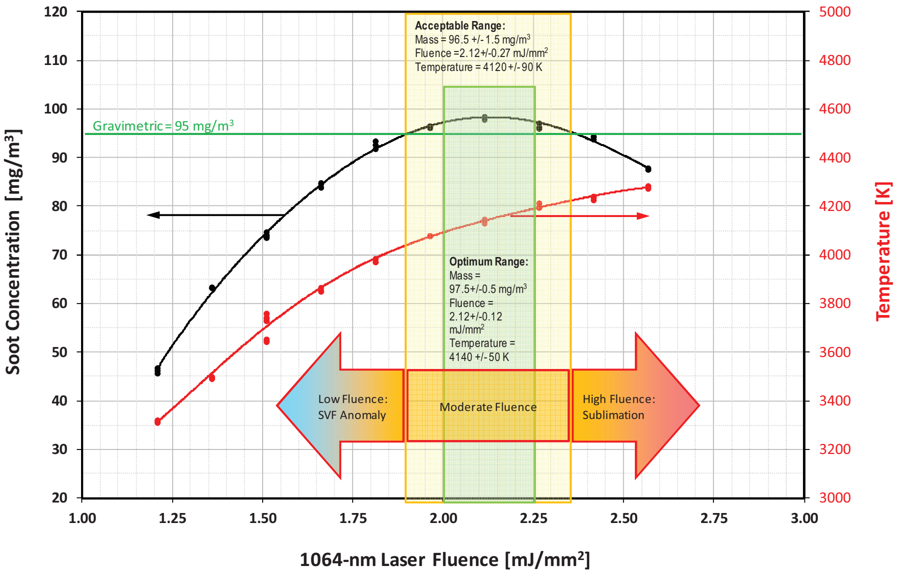

Fig. 1 Variation of soot concentration and temperature with laser fluence.

Effect of Laser Pulse Duration on Laser-Induced Incandescence of Soot

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The phenomenon of laser-induced incandescence (LII) has been utilized to develop versatile diagnostic techniques for measurements of combustion-generated soot concentration and primary particle size in many combustion applications. Many aspects in the implementation of LII have been investigated, such as the excitation laser wavelength and spatial profile, signal detection timing (prompt or delayed) and temporal width, and signal detection wavelengths. One potentially important parameter in LII practice that has received little attention in the LII community is the laser pulse duration. This is perhaps due to the fact that almost all the LII experiments conducted in the last two decades employed Q-switched, flashlamp pumped Nd:YAG lasers as the light source, which all provide a similar pulse duration in a narrow range between 5 and 10 ns FWHM. Few LII experiments were conducted using a much long laser pulse (microsecond) or much a shorter one (picoseconds). Picosecond laser pulses were shown theoretically to provide advantages over nanosecond ones in the determination of primary particle size distribution using low-fluence LII in flames at elevated pressures.

In this study the effect of laser pulse duration on the temperature histories of primary soot particle of different sizes was numerically investigated in both low- and high-fluence regimes under conditions of a typical atmospheric pressure laminar diffusion flame. The laser pulse durations considered in this study vary from 100 ps to 6 ns FWHM. Such laser pulse durations correspond to those of a typical Ti:Sapphire pulsed laser operated at 780 nm. Such laser is capable of generating laser pulses with duration from picoseconds to nanoseconds, depending on if the regenerative amplifier is seeded by the femtosecond oscillator.

Numerical results show that at a fixed laser fluence in the low-fluence regime with decreasing the laser pulse duration the effect of heat conduction on the peak soot temperature is suppressed. Under the conditions considered here heat conduction lowers the peak soot temperature of a 30 nm soot particle by about 70 K when the laser pulse is 6 ns FWHM. As the laser pulse duration decreases, the differences in the peak temperatures reached by particles of different sizes become smaller, due to the effective separation of the volumetric heating process and the surface dependent heat conduction cooling. In high-fluence regime, the peak soot temperature increases more significantly with decreasing the laser pulse duration. There seem no advantages using a shorter laser pulse than the commonly employed 6 ns FWHM one in the high-fluence regime.

In summary, a shorter laser pulse offers advantages over the typical 5 to 10 ns duration one in some low-fluence LII applications where it is desirable to suppress the effect of heat conduction, such as particle size determination in high pressures and evaluation of E(m). However, it does not seem to offer advantages in high-fluence regime.

Relationship between LII Signal and Soot Volume Fraction – Effect of Primary Particle Diameter Polydispersity

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LII has been increasingly applied to measure the concentration and size of nano-particles, such as combustion generated soot in flames, in-cylinder, and engine exhaust, black carbon in the atmosphere, and other synthesized nano-particles. The basis of LII concentration measurement is the assumption that the LII signal is proportional to the particle volume fraction, while the principle of LII particle sizing is the particle size dependence of heat conduction cooling after the laser pulse. In this work, the discussion is focused on measurement of volume fraction of combustion generated soot using LII.

Two approaches have been developed to conduct quantitative LII measurements of the soot concentration. The conventional method detects the LII signal at one wavelength in the visible and seeks a calibration constant using a known source of particle concentration, i.e., $S_{LII} = C \times f_v$. The second one is the more recently developed two-color LII or auto-compensating LII. In this method, absolute LII intensities are detected at two wavelengths in the visible spectrum to infer the soot temperature based on the pyrometry principle. This method does not require a known particle source to arrive at a calibration constant. However, it requires the knowledge of both relative and absolute values of E(m) at the two detection wavelengths, which represents the main uncertainty of the two-color LII method. The calibration constant C in the conventional LII method in general is not constant under conditions other than those of the calibration. Since soot temperature is determined in the two-color LII method, the two-color LII method can be viewed as a special version of the conventional LII method in which the calibration constant is obtain *in situ*.

It is shown numerically that for a polydisperse primary soot particles the soot temperature derived in two-color LII is biased towards the temperature of those larger and hotter particles. As smaller particles cool faster than larger ones, smaller particles gradually 'disappear', leading to a decrease soot volume fraction determined by the two-color LII. Based on numerically results, the relationship between LII signal and soot volume fraction can be summarized as:

(1) It is linear in the two-color LII during and shortly after the laser pulse in the low-fluence regime
(2) It is linear in the two-color LII only briefly around the peak of the laser pulse in the high-fluence regime
(3) It is in general non-linear in the conventional LII method

Novel soot volume fraction measurement through ratio-pyrometry and absolute light calibration

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Novel ratio pyrometry and absolute light calibration methods have been developed to obtain soot temperature and volume fraction in axisymmetric flames. A consumer digital single lens reflex camera has been fully characterized and utilized as a pyrometer. The incandescence from soot was imaged at the three wavelengths of the camera's color filter array (CFA). Temperatures were calculated by two-color ratio pyrometry using a lookup table approach. While temperatures can be extracted from color ratios, soot volume fraction requires an absolute light calibration of the detector. The absolute light intensity calibration was provided by a flame-heated S-type thermocouple. The spectral emissivity of S-type thermocouple wires (Pt and Pt-10% Rh) was measured in the visible range. The measured spectral emissivity, temperature, and diameter of the heated thermocouple wires allow them to serve as a light source with spectral radiance that can be calculated by Planck's law. Soot volume fraction measurements were carried out on four different flames with varying levels of soot loading. The results have been compared with previous LII results and excellent agreement has been achieved.

Modeling laser-induced incandescence of soot integrating spatial and temporal dependences of parameters involved in energy and mass balances

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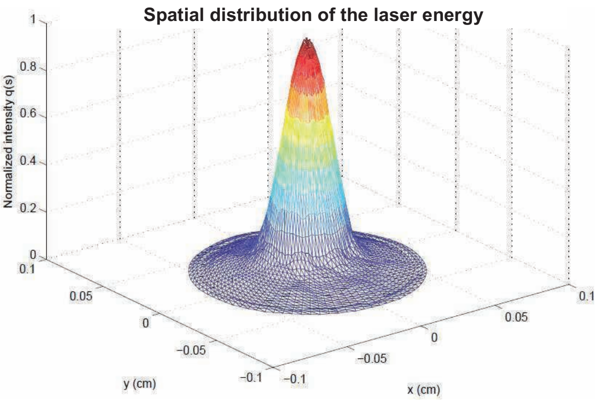
Laser-Induced Incandescence (LII) has become a widespread used technique for soot volume fraction and primary particle size determination in flames and exhaust gases. The correct interpretation of experimentally measured LII signals implies a detailed understanding of the physical mechanisms that control the LII phenomenon. It also needs, amongst other things, to thoroughly take into account the experimental parameters involved in the excitation process (especially the spatial and temporal profiles of the laser energy). Different models have been proposed recently in the literature to predict the temporal behavior of LII signals[1]. Nevertheless, except some recent works from Bladh et al.[2], only few works took into account the temporal and spatial characteristics of laser excitation sources presenting 2D inhomogeneous distributions.

In the present work, the experimentally monitored characteristics of an unfocused near-Gaussian laser beam have been considered as input data in our model (see the figure below). The spatial discretization of the mass- and energy-balance equations (based on the absorption (Q_{abs}), radiation (Q_{rad}), sublimation (Q_{sub}) and conduction (Q_{cond}) terms) has been carried out using the finite element method while the temporal discretization of these equations has been achieved following the Crank-Nicolson scheme. By this way, we obtain a matrix shape equations system in which the particles temperature and diameter (T_p and D_p, respectively) are the two unknowns. Such a 3D problem being non-linear, we solved it by using the Newton iterative method to obtain the evolution of T_p and D_p as a function of the time and of the space in each mesh of the excitation volume. The temperature dependence of parameters such as physical properties of soot has also been taken into account in the different terms used to obtain the mass- and energy -balance equations which allows determining the evolution of these properties for each time and spatial position.

A mesh sensitivity study has been carried out and the temporal evolution of Q_{abs}, Q_{rad}, Q_{sub}, Q_{cond}, T_p and D_p as a function of the space will be presented in this work which is still in progress. The spatial LII time decays that have been calculated by entering T_p and D_p into the Planck function integrated over a given range of wavelengths will be presented and potentially confronted with experimental data obtained using such a laser profile.



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EVOLUTION OF THE LASER INDUCED INCANDESCENCE SIGNALS OF SOOT PARTICULES MEASURED IN LOW-PRESSURE METHANE FLAMES

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The understanding of soot formation mechanisms in flames, and more specifically the nucleation process, is still under debate. To deal with this crucial step, low pressure laminar flames are particularly well suited because of the large reaction zone, offering the possibility of examining the early soot formation zone. The number of soot particles is however much lower than at atmospheric pressure, requiring the use of sensitive techniques such as Laser Induced Incandescence (LII).

In this work, we have used the LII technique to probe soot particles formed in various low-pressure premixed methane/oxygen/nitrogen flames stabilised for different equivalent ratio (Φ = 2.32, 2.05, 1.95) and pressure (P = 18.66 kPa (140 torr) and 26.66 kPa (200 torr)). Heating of the particles has been achieved by using the 1064 nm excitation wavelength of a YAG laser, the energy profile of which has been shaped as top hat. Temporal LII signals were measured by a photomultiplier whereas we complementary used an intensified CCD camera coupled to spectrometer in order to record the associated emission spectra. Measurements have been done for different heights above the burner (HAB), included the very beginning (nucleation step) of the soot formation processes in the flames.

By this way, we observed significant and surprising differences, mainly concerning the evolution of the temporal signal according the flame height, between the lowest equivalent ratio (Φ=1.95 and 2.05) and the reference flame (Φ=2.32). While this last flame is characterised by the increase of the LII decay with HAB, corresponding to the increase of the particles size as expected, no such evolution is observed for the two other ones. In these conditions, the temporal LII decays remain constant for all the heights above the burner, therefore questioning about the nature of the formed species. As a consequence, examination has been focused on those flames including fluence curves, measurement of relative volume fraction profiles and spectral analysis.

Influence of LII on Soot Optical Properties in Reference Flames

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When analyzing LII emission signals, it is typically assumed that the optical properties of soot are not affected by this rapid heating. However, from the literature it is known that the laser irradiances typical of 'plateau LII' can lead to significant modification of soot particles and even the formation of new particles [1,2]. When more moderate laser fluences are used, morphological changes are not observable via high resolution transmission electron microscopy; however, there is still evidence that the heating permanently influences the optical properties of the soot [3,4]. Variation of soot optical properties during or immediately after laser heating would have impacts on the interpretation of LII signal which should be accounted for in the theory in order to accurately use the emission data.

To study the optical properties of laser heated soot, we have monitored the extinction coefficient of soot aerosols within the standard Gülder and McKenna burners as a function of time while simultaneously heating the aerosol with laser pulses typical of LII. Extinction coefficient measurements were made at wavelengths of 405, 488, 632, and 804 nm and for a range of 1064 nm pulsed laser fluences.

We present a rich database of normalized extinction measurements which give clues into the complex consequences of rapid laser heating of soot aerosols. Normalized extinction coefficients show an enhancement of the propensity of soot to absorb light over the time interval of the laser heating. A partial relaxation of this enhancement is evident on the soot cooling time frame suggesting that the enhancement is in part due to a temperature based phenomena such as particle expansion or temperature dependent optical properties. A sustained residual enhancement is observed in the McKenna soot data, indicative of a permanent change to the soot optical properties, possibly due to graphitization. The variation of the normalized extinction coefficient in the McKenna burner diminishes with decreasing probe wavelength. This relates to the presence of non-soot material which absorbs light in the UV wavelengths, but is not heated by the 1064 nm laser. For the higher soot concentrations of the Gulder burner, heat transfer to the gas phase leads to a gas temperature change and expansion which decreases the attenuation propensity of the medium. The soot is more efficiently heated than the McKenna soot, with sublimation initiated at lower fluences and greater sublimation at a given fluence. This suggests a higher refractive index absorption function, $E(m_\lambda)$ for the Gulder soot. Normalized extinction coefficient measurements at 405 nm in the Gulder flame at very high fluences demonstrate that the materials vaporized from soot reform into species which are capable of absorbing 405 nm radiation, thus masking the sublimation effect on normalized extinction coefficient.

Both reversible and non-reversible changes to soot's ability to attenuate light have been demonstrated in McKenna and Gulder flame soot. These variations should be further quantified and incorporated into LII emission interpretation theory.

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Evaluation of particle sizes of iron-oxide nano-particles in a low-pressure flame-synthesis reactor by simultaneous application of TiRe-LII and PMS

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Laser-induced incandescence (LII) has become a common method for in-situ analysis of particle size and visualization of particle volume fractions predominantly for soot diagnostics in a wide range of applications. Besides lower signal strength due to less strongly absorbing material and lower heat-up temperatures, one of the main challenges when applying LII to non-carbon nanoparticles is the poor data base of relevant particle thermophysical properties, e.g., heat conduction, accommodation coefficients, vaporization enthalpy and high-temperature chemistry for describing particle cooling due to convection, vaporization, and other effects. In the present work the measured laser-induced emission signals from flame-synthesized iron oxide (Fe_2O_3) nanoparticles were evaluated in terms of particle sizing by using a modified version of the TiRe-LII model developed by Kock et al. [1].

Iron oxide nanoparticles were synthesized in a rich, premixed H_2/O_2/Ar low-pressure (30 mbar) flat flame in a low-pressure flame reactor that was doped with ppm-levels of $Fe(CO)_5$ as precursor material. By moving the burner relative to the fixed measurement location (determined by either the laser beam or a sampling nozzle for the particle mass spectrometer (PMS), respectively) the particle residence time in the reactor can be varied. The particles were heated by a frequency-doubled Nd:YAG laser and time-resolved LII-signal traces were recorded perpendicular to the beam axis by a two-color detection unit equipped with narrow band-pass filters with center-wavelengths at 500 and 700 nm, respectively, in front of two high-speed photomultipliers with integrated amplifiers. Additional to the time-resolved measurements, LII signals were detected spectrally-resolved using a spectrometer with an intensified CCD camera. The PMS with a molecular-beam sampling system was attached to the burner chamber for simultaneous particle sizing.

To determine a phenomenological evaporation heat flux term in the energy balance, temperature decay curves obtained by two-color pyrometry were fitted through variation of a parameterized form of the particle evaporation term. With these evaporation parameters, LII-signal traces were evaluated in terms of particle size. The obtained size parameters were verified by corresponding PMS measurements for the same flame conditions. In addition, it was possible to calculate the energy accommodation coefficient α_T of the present particle material at several experimental conditions. Emission spectra taken right after laser heating did not vary significantly in shape as a function of laser fluence.

The combination of TiRe-LII and online molecular beam particle sampling with subsequent particle mass spectrometry in low-pressure flames is a promising approach for fundamental research on the characteristics of LII of various nanoparticle materials.

Preference: Poster Presentation

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Comparison of different techniques for measurement of soot and PM emission from Diesel engine

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Here we present the comparison studies between different techniques for measurement of soot and particulate matter (PM) emissions from passenger car Diesel engine. The compared techniques include a filter paper type smoke meter, photo-acoustic spectrometer, opacimeter, differential mobility spectrometer and laser induced incandescence. We mainly focus our study to static and dynamic transient measurements tests from the location position closer to the actual combustion event - downstream of the turbine, position characterised by the higher temperature and higher pressure of the emission gas, than the standard measurement position, in the tailpipe of the exhaust manifold. The main task is to reveal the most accurate and sensitive method for fast soot and PM emission measurement for this particular measurement position. The issue of accuracy reliability of measured emission response and understanding of variances in measured soot emission due to different applied techniques can help to minimise the soot and a particulate matter emissions from diesel engines and to meet the future European emission standards.