6th International Workshop and Meeting on

Laser-Induced Incandescence: Quantitative Interpretation, Modeling, Application

June 8 - 11, 2014, Ven, Sweden

Program

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6th international Workshop and Meeting on Laser-Induced Incandescence

Program

Sunday, June 8, 2014

17:00 – 19:00	Registration at Backafallsbyn
18:00 – 20:00	Buffet at Backafallsbyn

Monday, June 9, 2014

8:00	Registration, Setting up of posters		
8:30	Welcome		
8:40	**Oral session 1**	8:40	Talk 1.1 - Advances in Modeling Laser-Induced Incandescence from Carbonaceous Particles; H. A. Michelsen
		9:00	Talk 1.2 - Influence of the internal multiple scattering on the absorption and scattering properties of soot fractal aggregates; J. Yon, F. Liu
		9:20	Talk 1.3 - An experimental and numerical study of gas heating by heat conduction from laser heated soot in a diffusion flame using LII and CARS; N.-E. Olofsson, E. Nordström, J. Simonsson, H. Bladh, P.-E. Bengtsson
		9:40	Talk 1.4 - Gas dynamics of sublimated species in high-fluence laser induced Incandescence; F. Memarian, F. Liu, K.A. Thomson, D.R. Snelling, G.J. Smallwood
		10:00	Talk 1.5 - Effects of volatile coatings and soot morphology on laser induced Incandescence; R. Bambha, M. Dansson, P. Schrader, H. A. Michelsen
10:20	**Coffee**		
10:50	Poster advertisement session		
12:00	**Lunch**		
13:20	**Oral session 2**	13.20	Talk 2.1 - Quantifying the Thermal Accommodation Coefficient for TiRe-LII Analysis of Iron Nanoparticles; T. A. Sipkens, N. R. Singh, K. J. Daun, N. Bizmark, M. Ioannidis, J. T. Titantah, M. Karttunen
		13:40	Talk 2.2 - Effects of organic carbon fraction and removal of organic material on light extinction by laser-heated soot; M. Saffaripour, K.P. Geigle, K. Thomson, D.R. Snelling
		14:00	Talk 2.3 - On the Way to In-Cylinder 2D Time-Resolved LII measurements; A. Maier, A. Dreizler
		14:20	Talk 2.4 - Photoacoustic Soot Measurement: Comparison with LII; J. Black, G. S. Humphries, J. Dunn, M. Lengden, I. S. Burns
		14:40	Talk 2.5 - Assessment of soot particle-size imaging with LII at Diesel engine Conditions; E. Cenker, K. Kondo, G. Bruneaux, T. Dreier, T. Aizawa, C. Schulz
15:00	**Coffee**		
16:00	Visit to the Tycho Brahe museum		
18:00	**Dinner**		
19:30 - 23:00	Poster session		

Tuesday, June 10, 2014

8:30	**Discussion session 1**	8:30	**Measurements in LII standard flames** ***Chair:*** *Klaus-Peter Geigle, DLR Stuttgart, Germany* Overview of LII relevant information measured in the LII standard flames (focus) and trend studies, i.e. change of burner material, measurement location in flame, or other parameters.
		9:15	**Determination of key parameters for LII** ***Chair:*** *Stefan Will, LTT Erlangen, Germany* Experimental/numerical/theoretical contributions to the determination of key parameters for LII such as accommodation coefficient, complex index of refraction/absorption function, etc.
10:00	**Coffee**		
10:30	**Oral session 3**	10:30	Talk 3.1 - Experimental Investigation of the impact of imposed air inlet velocity oscillations on Soot Formation and Oxidation using an advanced 2-Colour-TIRE-LII; A. Aleksandrov, H. Bockhorn, R. Suntz
		10:50	Talk 3.2 - Correlated laser-induced fluorescence of PAH and laser-induced incandescence to visualize soot inception in turbulent flames; K.P. Geigle, W. O'Loughlin, W. Meier
		11:10	Talk 3.3 - Soot size and concentration in combusting sprays at high gas pressures and elevated temperatures estimated by optical methods; R. Ochoterena, M. Andersson, S. Andersson
		11:30	Talk 3.4 - Laser-induced incandescence (LII) measurements on gas-borne silicon Nanoparticles; R. Mansmann, T. Dreier, H. Wiggers, C. Schulz
		11:50	Talk 3.5 - Aerosol mass spectrometry of refractory black carbon containing particles; T.B. Onasch, E.C. Fortner, P. Massoli, L.R. Williams, A.T. Lambe, A.M. Trimborn, J. T. Jayne, P. Davidovits, and D.R. Worsnop
12:10	**Lunch**		
13:30	**Discussion session 2**	13:30	**LII of emitted and ambient soot** ***Chair:*** *Kevin Thomson, NRC, Ottawa, Canada* Experimental/numerical/theoretical activities, issues, and best practices of LII applied to non-volatile particulate matter (soot, black carbon) emitted from combustion processes as an exhaust or after it has evolved in the atmosphere.
		14:15	**Combined techniques** [S. de Iuliis] ***Chair:*** *Silvana de Iuliis, CNR-IENI, Milan, Italy* Experimental and numerical approaches on the combination of LII with optical (scattering, emission, absorption, etc...) and/or non-optical diagnostics (TEM analysis, photoacoustic, etc). Burning issues to gain more insight on the comprehension of the technique and on the soot optical properties.
15:00	**Coffee**		
15:30	Free afternoon		
18:00	Poster session		
19:00	**Dinner**		
21:00 – 23:00	Poster session		

Wednesday, June 11, 2014

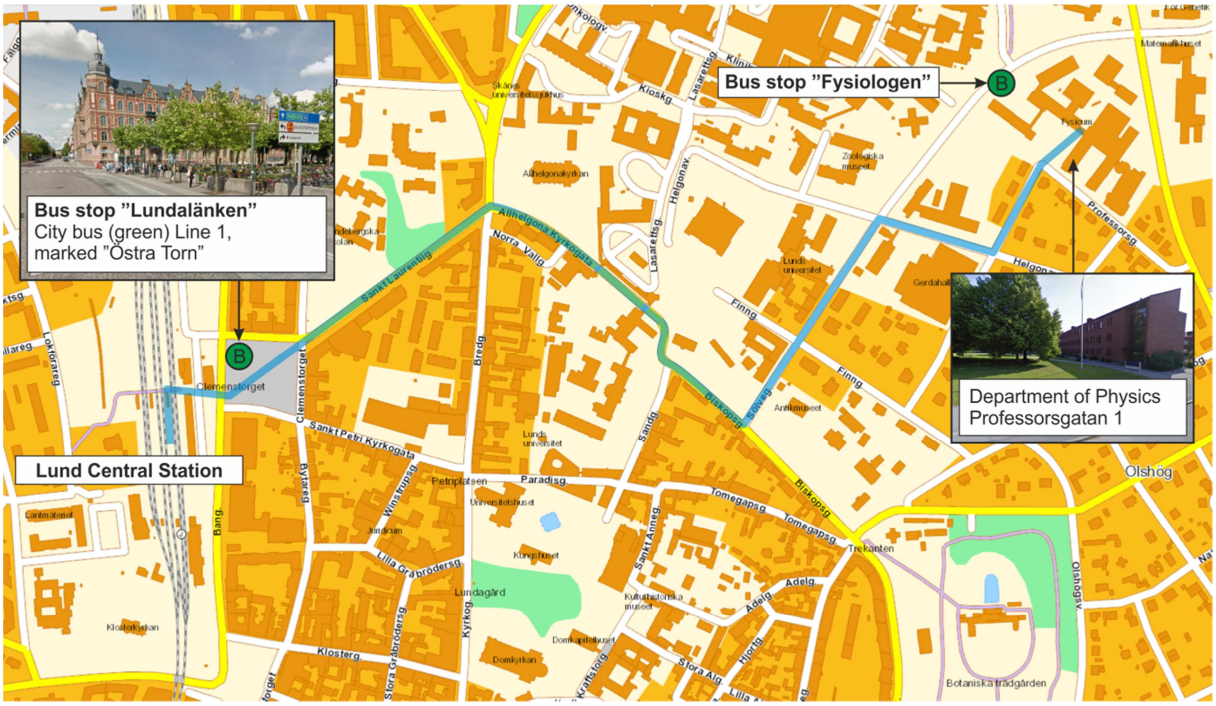
8:00	Poster session (9:00 - 10:00: Advisory committee closed meeting)		
10:00	**Coffee**		
10:30	**Discussion session 3**	10:30	**Non-soot LII** ***Chair:*** *Christof Schulz, IVG and CENIDE, University of Duisburg-Essen, Duisburg, Germany* Fundamental experiments and simulation (optical properties, accommodation coefficients, fluence dependence) on non-soot particles/new application of LII to non-soot particles/LII combined with other laser-induced emission signals or scattering, etc.
		11:15	**LII modeling** ***Chair:*** *Hope Michelsen, Sandia National laboratories, CA, USA* Theoretical modeling of the LII process. The latest developments with regards to the model descriptions of the heat-up process involved in LII will be discussed.
12:00	Summary and closure of workshop		
12:30	**Lunch**		

Thursday, June 12, 2014

There will be a possibility of a guided tour at the division of Combustion Physics in Lund at 08:30. Optionally more tours can be arranged later the same day.

Directions:

Take a train towards Malmö from the Landskrona train station. All trains make a stop in Lund. At the Lund central station it is a 15 minutes' walk to the Department of Physics. Buses depart 4 times an hour from the bus stop at Clemenstorget called "Lundalänken". Take green city bus, Line 1, marked "Östra Torn". If you want to grab a Taxi, the address is Professorsgatan 1.



Posters

Number	Title
1	In situ analysis of the nanoparticle formation in the gas phase during carbon nanotube synthesis; A. Dichiara, Y. Ma, L. Zimmer, J. Bai
2	Mo nanoparticle sizing by Ti-Re LII and TEM; A. Eremin, E. Gurentsov, M. Yurischev
3	Soot measurements in premixed high-pressure flames using light emission, TiRe-LII, laser extinction, and TEM-sampling; M. Leschowski, T. Dreier, C. Schulz
4	LII in an Aero-Engine Exhaust Using a Low Peak Power Fibre Laser; J. D. Black, D. McCormick, Y. Feng
5	Spectrally- and temporally-resolved laser-induced incandescence (LII) on gas-borne silicon nanoparticles with varying laser fluence; J. Menser, T. Dreier, C. Schulz
6	Quantitative Measurements of Soot Volume Fraction Using Planar LII in Diesel Spray Combustion under Diesel Engine Conditions; Y. Gao, F. Liu, X. He, F. Liu, L. Zheng, J. Wang
7	Application of Planar Laser Induced Incandescence in Turbulent and Sooting Flames: The Influence of Radiation Trapping and Beam Steering; Z. T. Alwahabi, Z. W. Sun, G. J. Nathan, B. B. Dally
8	Real-time Capable Characterization of Soot Nanoparticles by Wide-Angle Light Scattering (WALS); F. Huber, M. Altenhoff, S. Will
9	Approach to standardize a spray-flame nanoparticle synthesis burner; J. Menser, S. Kluge, T. Dreier, C. Schulz
10	Soot optical properties investigation by two-color laser-induced incandescence measurements; F. Migliorini, S. De Iuliis, G. Zizak
11	Determination of small soot particles in the presence of large ones from time-resolved laser-induced incandescence; E. Cenker, G. Bruneaux, T. Dreier, C. Schulz
12	Sensitivity analysis for in-cylinder soot-particle size imaging with laser-induced Incandescence; E. Cenker, G. Bruneaux, T. Dreier, C. Schulz
13	Assessment of soot particle-size imaging with LII at Diesel engine conditions; E. Cenker, K. Kondo, G. Bruneaux, T. Dreier, T. Aizawa, C. Schulz
14	Soot volume fraction measurement by extinction and Laser Induced Incandescence in a wood-fired boiler under varying boiler conditions; S. Bejaoui, E. Therssen
15	Measurement of soot temperature, concentration and cooling rate; and bulk fluid temperature using modulated laser induced incandescence; D.R. Snelling, K.A. Thomson, R. A. Sawchuk, G.J. Smallwood
16	Probing the smallest soot particles in low-sooting premixed flames using laser-induced incandescence; H. Bladh, N.-E. Olofsson, T. Mouton, J. Simonsson, X. Mercier, A. Faccinetto, P.-E. Bengtsson, P. Desgroux
17	Mini-CAST as a Cold Soot Source for Studying Optical Properties of Well Characterized Carbonaceous Particles; S. Török, A. Eriksson, J. Simonsson, N-E. Olofsson, J. Pagels, H. Bladh, P-E. Bengtsson
18	Studies of optical and physical properties of soot in premixed flat flames using laser-induced incandescence, elastic light scattering and extinction; J. Simonsson, N.-E. Olofsson, S. Török, H. Bladh, P.-E. Bengtsson
19	Effect of primary particle polydispdersity on the absorption cross section of soot aggregate and the implications to the soot absorption function derived from low-fluence LII; F. Liu, J. Yon
20	Comparison of LII and Extinction Measurements of Soot Volume Fraction in Turbulent Jet Flames; C. R. Shaddix, J. Zhang
21	Soot Measurements in Counterflow Non-premixed Flames using Laser Induced Incandescence: Soot Volume Fraction, Particle Size, and Number Density; B.G. Sarnacki, H.K. Chelliah

Abstracts

Oral presentations

Advances in Modeling Laser-Induced Incandescence from Carbonaceous Particles

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This talk introduces a new model that describes the temporal behavior of the temperature and size of carbonaceous particles heated by a continuous wave or pulsed laser. The model uses calculated particle temperature and size to predict the time evolution of laser-induced incandescence signal, elastic light scattering, and extinction, accounting for changes in the physical and optical properties of the particle induced by laser heating. Particles are heated by laser absorption, surface oxidation, and annealing and are cooled by conduction to the surrounding atmosphere, radiative emission, thermionic emission, and sublimation of molecular carbon clusters. Particles lose mass in the model by sublimation, non-equilibrium desorption, and oxidation. The model allows annealing of particle fine structure to reduce conductive cooling rates and increase absorptive heating rates and accounts for effects of soot maturity on thermal accommodation, particle density, and emissivity. The model is validated with data collected from soot in two regions of a co-flow diffusion flame using pulsed-laser excitation at 532 and 1064 nm over a wide range of laser fluences. It reproduces measured LII signal and particle temperature temporal profiles, including the observation that temperatures of laser-heated soot reach, but do not exceed, the sublimation temperature of C_2 (4459 K) at high laser fluences. It also reproduces measured extinction at 532 and 1064 nm.

Keywords: LII, soot, modeling, thermal accommodation, absorption

Influence of the internal multiple scattering on the absorption and scattering properties of soot fractal aggregates

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Light extinction, light scattering and Laser induced incandescence are established and popular optical and in-situ techniques for measurements of soot particle size and volume fraction.
Due to the complex morphology of these particles and the computation time needed for the evaluation of their accurate radiative properties, the optical measurements are usually interpreted by the Rayleigh Debye Gans theory for fractal aggregates (RDG-FA). Even if this simple theory is often considered reliable given the experimental uncertainties[1]; several important hypotheses are made including that radius of the primary sphere has to be much smaller than the wavelength (Rayleigh regime), each primary spheres is supposed to be exposed to the incident light source and multiple scattering inside the aggregate is neglected.
In a recent study, we suggested to incorporate the multiple scattering effects in the RDG-FA[2]. Firstly, a brief descriptions of the methodology used is proposed in the present communication.

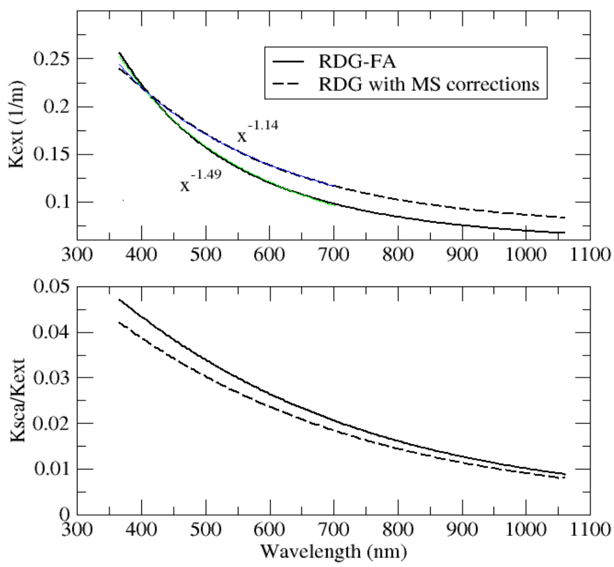

Fig. 1 Impact of aggregate internal multiple scattering on the evaluation of the extinction spectra.

After that, the impact of multiple scattering on the theoretical evaluation of the absorption and scattering cross sections will be presented. For example, Fig. 1 presents the impact of accounting for the multiple light scattering inside the aggregates on the extinction spectra calculated in case of Diesel soot particles[3]. It is shown that multiple scattering softens the decrease of the light extinction as a function of the wavelength in the near UV spectrum. The multiple scattering also reduces contribution of the total scattering in the extinction coefficient.
The impact of these results on the LII measurement will be discussed. The impact of the multiple scattering effects on angular light scattering will also be presented showing that measurements can be misinterpreted by the RDG-FA theory as a function of the chosen wavelength. For example, we show that the fractal dimension determined by angular scattering measurements at 266 nm can lead to an over prediction of the fractal dimension by a factor higher than 8%.

Keywords: Soot, Optical properties, RDG-FA, multiple scattering.

[1] R. K. Chakrabarty, H. Moosmüller, W. P. Arnott, *et al*, *Appl. Opt*., **46**, 6990-7006 (2007).
[2] J. Yon, F. Liu, A. Bescond, et al, *J QuantSpectroscRadiatTransfe*, **133**, 374-381 (2014).
[3] J. Yon, R. Lemaire, E. Therssen, *et al.*, *Appl. Phys. B*, **104**, 253-271 (2011).

An experimental and numerical study of gas heating by heat conduction from laser heated soot in a diffusion flame using LII and CARS

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In laser-induced incandescence (LII) the strong light absorption by soot particles is used to heat the particles to temperatures above 4000 K by high energy laser pulses. Consequently, when the heated soot cools down, the ambient gas temperature will increase by heat conduction from the particles. This local gas heating is an effect that has received minor attention so far in the diagnostic community. Clear indications of a local gas heating effect were found by Snelling et al.[1] in a study from 2009, where two-color pyrometry was used to observe that the soot particles cool down asymptotically towards a significantly higher temperature than the initial flame temperature.

In the present work, a pump-probe-type experiment was designed to measure the local gas heating effect in a particle-laden flame, see Fig. 1. A 1064-nm pulsed laser was used to heat the soot particles in an ethylene diffusion flame on a Gülder burner, see Fig. 2. To ensure homogenous heating of the soot, a top-hat spatial profile was used. The local gas temperature was probed by using a two-beam rotational coherent anti-Stokes Raman spectroscopy (CARS) setup. By changing the delay between the CARS-laser and the LII-laser, the local gas temperature could be measured on time scales from nanoseconds to milliseconds. The particle temperatures were simultaneously probed by two-color pyrometry, using a two-color laser-induced incandescence (2C-LII) setup. Measurements were conducted both at low laser fluences, leading to minor sublimation, and at high laser fluences, giving rise to major sublimation of the soot particles. The results show that laser heating of soot particles from flame temperatures to sublimation temperatures leads to local gas heating of ~100 K at a soot volume fraction of 4 ppm, in good agreement with theoretical predictions.

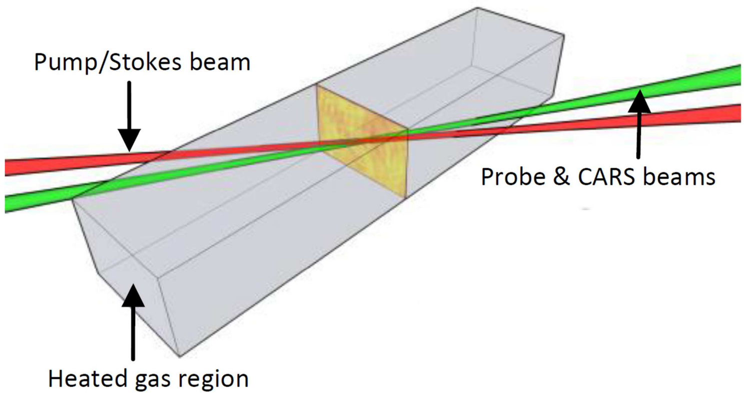

Fig. 1 A sketch of the measurement point, showing the crossing of the heating 1064-nm LII laser beam and the CARS laser beams. The gray box represents the heating laser with a top-hat profile and the CARS beams are indicated by arrows.

Fig. 2 The ethylene diffusion flame used in this study. Measurements were made at 42 mm height above the burner nozzle.

Keywords: Diffusion flame, soot, gas heating, gas temperature, rotational CARS

[1] D.R. Snelling, K.A. Thomson, F. Liu, G.J. Smallwood, Appl. Phys. B **96**, 657-669 (2009).

Gas dynamics of sublimated species in high-fluence laser induced incandescence

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Although significant progress has been made towards understanding many aspects of heat and mass transfer in LII over the last three decades, the physics of LII in the high-fluence regime (at soot temperatures beyond 3,800 K) remains relatively poorly understood. Under these conditions, current LII models significantly over-predict the soot temperature decay rate compared to those inferred from two-color pyrometry after the peak of the laser pulse[1]. Some LII researchers speculate that this discrepancy could be due to possible gas dynamics effects involving sublimed carbon nanoclusters, such as recondensation of sublimed nanoclusters or formation of shock waves[1].

Recent research has used transient Direct Simulation Monte Carlo (DSMC) to investigate how sublimed nanoclusters could influence the local gas dynamics and heat transfer from a laser-energized nanoparticle. In the initial study[2], a predetermined temperature decay curve was specified for the soot particle; the results confirmed that a back flow of sublimed species occurs due to intermolecular collisions close to the nanoparticle surface as indicated by a two-sided Maxwellian velocity distribution, as shown in Fig. 1; on the other hand no shock wave forms, even under extreme conditions. Subsequently, a new method of ensemble averaging was used to couple the transient DSMC simulation directly to the nanoparticle cooling model. The cooling curve of a single soot nanoparticle (both a single soot primary particle and single soot aggregate) is calculated as it undergoes laser absorption, conduction and sublimation heat losses. Fig. 2 shows a comparison between DSMC and LII heat transfer predictions and also shows the ratio of condensation to sublimation heat transfer predicted by DSMC. This figure implies that condensation of sublimed species on the soot nanoparticle from which they have originated is not significant enough to cause a large error in LII model predictions.

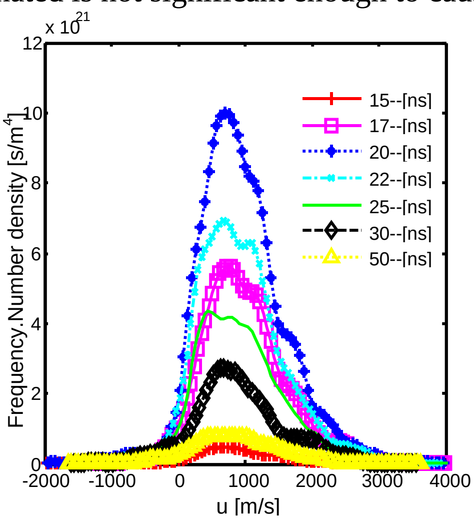

Fig. 1: Velocity distribution of sublimed carbon nanoclusters

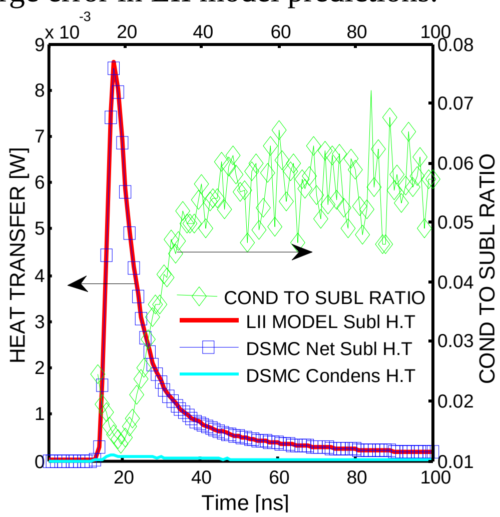

Fig. 2: Nanoparticle cooling decay determined using transient DSMC

Keywords: DSMC, Transient Heat Transfer, Sublimation, LII, Gas Dynamics

[1] D.R. Snelling, *Applied Physics B*, 96(4), 657-669 (2009)
[2] F. Memarian, K.J. Daun, *Numerical Heat Transfer, Part B: Fundamentals* 65(5), 393-409 (2014)

Effects of volatile coatings and soot morphology on laser induced incandescence

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Soot particles in exhaust plumes are often coated[1,2], and during subsequent atmospheric transport, additional coating can be formed. In a diesel exhaust stream, for example, particles can be comprised of as much as 50% volatile compounds.[3] These coatings can have a significant effect on the signal produced during measurements of laser induced incandescence (LII).[4] If the soot coatings are removed by thermal denuding, a permanent restructuring of the particle morphology can result that may also significantly alter the LII signal. The effects of particle coatings and morphology therefore need to be fully understood in order to apply optical diagnostics under a wide range of conditions.

We have measured time-resolved LII from uncoated soot generated in a coflow diffusion flame and compared these measurements with LII from 1) soot coated with oleic acid and 2) soot coated with oleic acid and subsequently thermally denuded to remove the volatile coating. LII measurement were performed on soot that had been size selected using a differential mobility analyser and characterized with a scanning mobility particle sizer, centrifugal particle mass analyser, and transmission electron microscope. These results demonstrate differences in the LII from coated, uncoated, and restructured particles in terms of both the temporal evolution and dependence on laser fluence. Enhancement of absorption cross section due to coatings appeared to be negligible under the conditions we employed. An enhancement in LII signal of restructured particles appeared to have resulted from lower conductive cooling. This interpretation is supported by LII model results.

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.
(4) Bambha, R. P.; Dansson, M. A.; Schrader, P. E.; Michelsen, H. A. *Applied Phys. B* 2013, *3*, 343-358.

Keywords: LII, soot, coatings, atmospheric

Quantifying the Thermal Accommodation Coefficient for TiRe-LII Analysis of Iron Nanoparticles

T. A. Sipkens[1], N. R. Singh[1], K. J. Daun[1], N. Bizmark[1], M. Ioannidis[1], J. T. Titantah[2], M. Karttunen[1]
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[2]*Western University, 1151 Richmond St, London, ON, Canada, N6A 3K7*
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Desired format: Oral presentation

The thermal accommodation coefficient (TAC), α, which defines the energy transfer between the laser-energized nanoparticle and surrounding gas, must be known in order to extend time-resolved laser-induced incandescence to synthetic aerosols. The TAC can be inferred from TiRe-LII data if the nanoparticle sizes are known, e.g. through electron microscopy[1], while the nanoparticle size and TAC can be inferred simultaneously if evaporation plays an important role in nanoparticle cooling[2,3]. More recently, molecular dynamics has been used to predict the TAC[4]. Unfortunately, there remains considerable uncertainty surrounding the value of the TAC for synthetic nanoparticles.

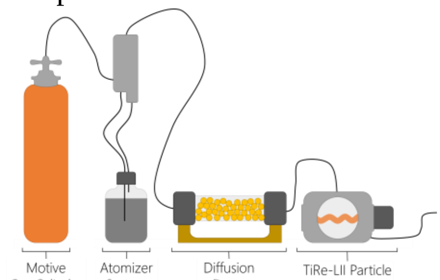

Fig. 1. Schematic of the experimental apparatus.

In order to quantify the TAC for aerosols of iron nanoparticles, and to elucidate the gas-surface scattering physics that underlies this parameter, TiRe-LII measurements were carried out on nanoparticles formed in solution by reducing ferrous iron (Fe^{2+}) with sodium borohydride ($NaBH_4$), capped with carboxymethylcellulose (CMC) to prevent agglomeration and oxidation. The nanoparticles were then aerosolized with a pneumatic atomizer using a variety of motive gases, dried with a diffusion drier, and then passed through an Artium 200 M instrument. In contrast to previous studies[1-3], nanoparticle synthesis is independent of the motive gas, so the nanoparticle sizes should be consistent and any differences in the TiRe-LII data between motive gases should be due to differences in the TAC. As evaporation contributes to the nanoparticle cooling rate, the TAC and nanoparticle size can be inferred simultaneously from the TiRe-LII data[1,2,4]. Inferred nanoparticle sizes are generally consistent with sizes inferred through electron microscopy and dynamic light scattering. The TACs for Fe/He, Fe/Ne, and Fe/Ar increase monotonically with gas molecular mass, while the TACs for most of the polyatomic gases are lower since energy is accommodated less efficiently into the rotational mode of the gas molecule. The exception to this trend is Fe/CO, which was the last measurement carried out in the series. This outlier could be due to an observed buildup of CMC on the sample chamber windows, or oxidation and agglomeration within the iron nanoparticle solution.

Aerosol	exp. (pres. study)		α, exp. (lit)	α, MD
	d_p (nm)	α		
Fe-He	45	0.01	0.01[1]	0.10[4], 0.07
Fe-Ne	36	0.08	-	-
Fe-Ar	38	0.14	0.1[1], 0.13[2]	0.23[4], 0.13
Fe-N_2	38	0.10	0.13[2]	0.08
Fe-CO	55	0.31	0.2[1]	-
Fe-N_2O	36	0.08	-	-
Fe-CO_2	34	0.09	-	-

The experiments were complimented with molecular dynamics simulations of He, Ar and N_2 interacting with an iron surface. Trends in MD-derived TACs are consistent with experimentally-observed values.

[1] A Eremin, E Gurentsov, C Schulz, *J. Phys. D*, 41 (2008) 055203.
[2] B.F. Kock, C. Kayan, J. Knipping, H.R. Orthner, P. Roth, *Proc. Combust. Inst.*, 30 (2005) 1689.
[3] T.A. Sipkens, et al., *Appl. Phys. B* (2013).
[4] K. J. Daun, T. A. Sipkens, J. T. Titantah, M. Karttunen, *Appl. Phys. B*, 112 (2013) 409.

Keywords: iron, thermal accommodation coefficient, molecular dynamics

Effects of organic carbon fraction and removal of organic material on light extinction by laser-heated soot

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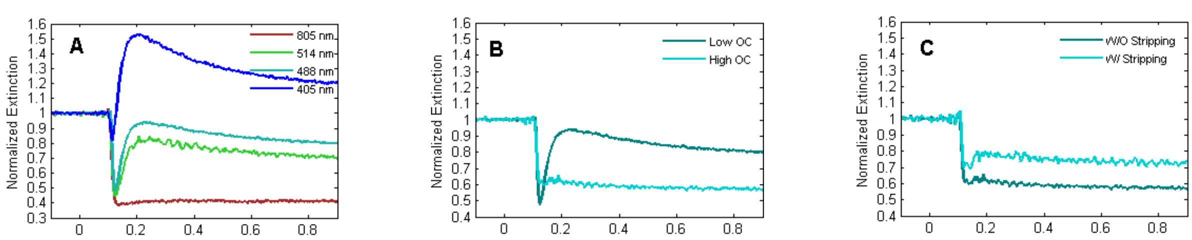
To investigate the impact of organic carbon fraction and removal of organics on light interaction with soot particles during a rapid laser heating event, continuous wave (CW) light extinction at several wavelengths is performed on a cooled soot plume heated by a 1064 nm laser pulse. A quenched propane/air flame is used to generate a steady source of cooled soot with different amounts of organic fraction (4% and 58% by mass [1]). The results for untreated soot are compared to the results for soot particles stripped of organics in a catalytic stripper. The present work improves upon our previous study [2] on untreated cold soot particles.

For untreated soot with low organic content (4%), an enhancement of light extinction (by up to 12%) occurs immediately after the laser pulse (~0.1 μs) that is attributable to some degree of graphitization, which is an irreversible process, as well as reversible and temperature dependent enhancement of the absorption coefficient of soot particles. This extinction increase is smaller at lower CW wavelengths, suggesting that the enhancement is caused by an optical property related effect. The increase of extinction is followed by a sharp decline (up to 62%) caused by soot sublimation and a rapid loss of material from the soot particles (Fig. A). The sublimation fragments produce secondary gaseous species that are strong absorbers of light at low wavelengths (such as C_3 Swings band that absorb strongly at 405 nm). Therefore, a steep rise of extinction which in some cases exceeds the initial extension of the unheated soot is observed at low CW wavelengths shortly after the start of sublimation (Fig. A).

Untreated soot particles with high organic content (58%) do not exhibit an initial rapid extinction enhancement. It is speculated that any enhancement due to graphitization is masked by the substantial contribution of organic material desorption. The sharp drop of extinction for these particles is caused mainly by desorption of organics rather than sublimation as carbon vapor, hence it begins at much lower IR laser fluences (as low as 0.4 mJ/mm^2). The secondary rise in extinction, which was observed after the start of sublimation for particles with low organic content, does not occur for organic-rich particles (Fig. B). It is speculated that the desorbed organic species are less energetic such that they do not form the highly absorbing secondary carbon fragments.

Removal of the organic material does not affect the extinction results significantly for soot particles containing a small amount of organics. However, the impact is substantial for particles with initially high organic content (see Fig. C). The steep decline of extinction at lower CW wavelengths is much smaller for stripped particles because of the significant loss of light absorbing organic material in the catalytic stripper. Secondary species are not produced in considerable amounts by the stripped particles either, indicating that the core material of the untreated particles is less graphitic and therefore less prone to sublimation as carbon fragments.



(A) Time-resolved attenuation profiles at four different CW wavelengths for untreated soot with low organic content; (B) Extinction profiles of soot particles with high and low organic carbon levels at 488 nm; (C) Extinction profiles of untreated and stripped organic-rich particles at 488 nm. The IR laser fluence used for all three figures is 5.45 mJ/mm^2.

Keywords: cold soot LII, time-resolved extinction, organic carbon fraction, stripped soot

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2 Thomson et al., Appl. Phys. B 104, 307 (2011)

On the Way to In-Cylinder 2D Time-Resolved LII measurements

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In order to meet future soot emission regulation, there is a need to understand the particulate formation mechanisms in combustion engines. Information about the in-cylinder particle size distribution might be the key to identify and minimize particulate formation. The application of Time-Resolved Laser-Induced Incandescence (TiRe-LII) to measure the mean size of soot particles within the cylinder of a combustion engine has been reported in the literature[1]. The signals were recorded using photomultipliers and the measurements were thus limited to specific "points" in the combustion chamber. For stationary flames it is possible to assemble a two-dimensional size distribution by combining measurements carried out with an intensified camera at different gate timings, assuming the flame is stable[2]. However, an extension of this approach to combustion engines is hardly feasible due to the significant cycle-to-cycle fluctuations.

In this work, a measurement setup leading to a 2-dimensional map of the exponential decay coefficient of the LII signal is suggested and its application demonstrated for a stationary burner. A Gaussian light sheet is produced using a Nd:YAG laser at the fundamental wavelength of 1064 nm. The detector is a camera with an integrated beam splitter and eight independently controlled intensified chips. The first image of the sequence is recorded just before the firing of the laser pulse and thus sets the baseline soot luminescence. Seven additional images follow after the laser pulse with an illumination time of 50 ns each and a delay of 50 ns between them. Each image was corrected for differences of each chip using a white LED screen as reference. The exponential decay time τ ($I=I_0 \cdot e^{-t/\tau}$) is then calculated individually for a binned region of 9 pixels. For this first demonstration, well-characterized burners producing stable flames such as the McKenna and Gülder burners are used. In principle the extension of the technique to instationary processes such as combustion engines is straightforward. Given the fluctuation of the decay coefficient in Fig. 1 for an average of 30 images, it is obvious that the measurement setup has still to be optimized towards a higher signal-to-noise ratio, to be able to evaluate single-shot events.

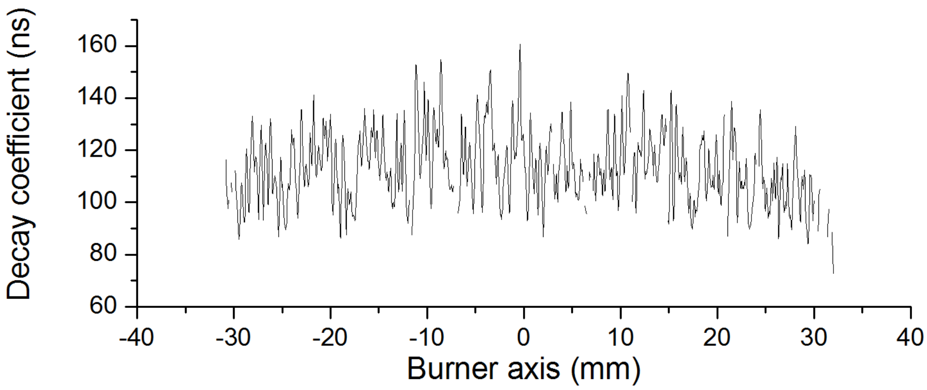

Fig. 1. Exponential decay coefficient across horizontal line at approximately 12 mm height above McKenna burner (equivalence ratio = 2.1, 30 images averaged before calculation of decay coefficient)

Keywords: 2D TiRe-LII

[1] B.F. Kock, B. Tribalet, C. Schulz and P. Roth, *Combustion and Flame* **147,** 79-928 (2006)
[2] R. Hadef, K.P. Geigle, J. Zerbs, R. Sawchuk and D.R. Snelling, *Appl. Phys. B* **3,** 395-408 (2013)

Photoacoustic Soot Measurement: Comparison with LII

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Here we present a photoacoustic detection method which shows promise for the in-situ measurement of soot in a premixed ethylene/air flame stabilised on a McKenna burner.

Photoacoustic detection is based on the generation of acoustic pressure waves by an optical energy source, such as a laser. Soot particles in the flame produce an acoustic wave by thermal release to the surrounding gas medium following heating.

For this work, acoustic waves are generated using a high power (30W), λ = 808nm, continuous wave diode laser to produce local heating due to absorption by soot in the flame. The laser intensity is modulated using an optical chopper, and the resulting acoustic signal is then detected using a Wolfson MEMS microphone and a lock in amplifier.

Early results acquired using the photoacoustic technique have shown a good match to those obtained from prompt laser induced incandescence (LII) measurements performed in the same flame using a pulsed 1064nm Nd:YAG laser.

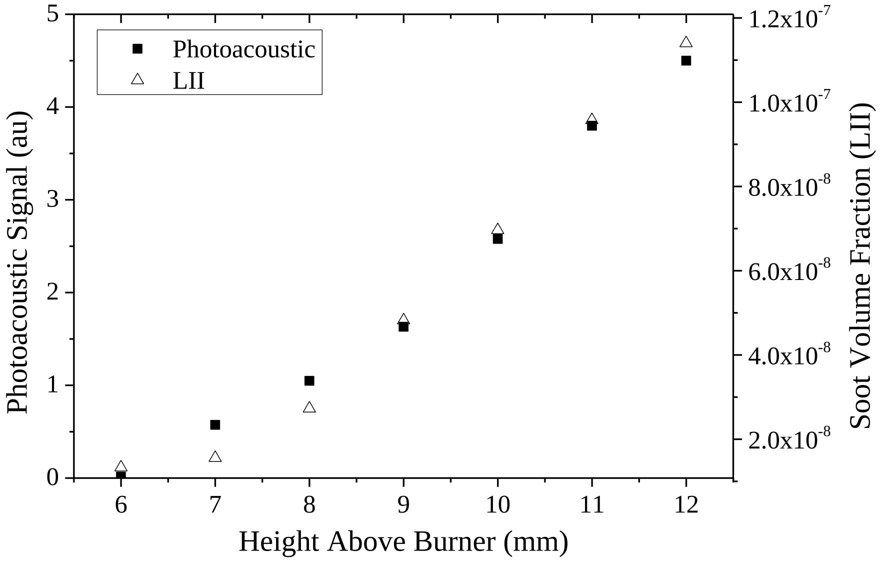

Fig. 1. Comparison of profile obtained from photoacoustic detection with 1064nm LII calibrated to soot volume fraction. These results were taken in a 2.1 equivalence ratio C_2H_4/Air premixed flame

The data in Figure 1 above shows a good match in the shape between the profile obtained using photoacoustic detection with soot volume fraction measurements obtained using extinction calibrated 1064nm LII..

The use of a photoacoustic detection scheme allows for a relatively simple experimental setup, removing the complex detection optics inherent with purely optical techniques (such as LII) using instead only a microphone and a lock in amplifier.

Keywords: Photoacoustic, Diode Laser, Soot.

Assessment of soot particle-size imaging with LII at Diesel engine conditions

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Two-time-step laser-induced incandescence (LII) imaging was performed in Diesel engine-relevant combustion to investigate its applicability for spatially-resolved measurements of soot primary particle sizes. The method is based on evaluating gated LII signals acquired with two cameras consecutively after the laser pulse and using LII modeling to deduce particle-size from the ratio of local signals. Based on a theoretical analysis, optimized detection times and durations were chosen to minimize measurement uncertainties. The dependence of LII particle-size imaging on the assumed boundary conditions is identified such as bath gas temperature, pressure, particle heat-up temperature, accommodation coefficients, and soot morphology. Various laser-fluence regimes and gas pressures are considered. Effects of laser attenuation are evaluated. Experiments were conducted in a high-temperature high-pressure constant-volume pre-combustion vessel under the Engine Combustion Network's (ECN) "Spray A" conditions at 61–68 bar with additional parametric variations of injection pressure, gas temperature and composition. The LII measurements were supported by pyrometric imaging measurements of particle heat-up temperatures. The results were compared to particle-size and size-dispersion measurements from transmission electron microscopy (TEM) of soot thermophoretically sampled at multiple axial distances from the injector. The discrepancies between the two measurement techniques are discussed to analyze uncertainties and related error sources of the two diagnostics. It is found that in such environment where particles are small and pressure is high, LII signal decay times are such that LII suffers from a strong bias towards large particles.

Keywords: Soot particle-size imaging, Diesel jet, ECN, TEM, pyrometry imaging

Experimental Investigation of the impact of imposed air inlet velocity oscillations on Soot Formation and Oxidation using an advanced 2-Colour-TIRE-LII

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Abstract
In gas turbine combustion in partially premixed mode or diffusion mode (e.g. "rich-quenched-lean" (RQL) concept) soot is formed intermediately which necessarily has to be oxidized before entering the exhaust. In spite of the achieved progress in the last 50 years[1,2], models for soot formation and oxidation that are able to predict soot emission from gas turbines under highly turbulent conditions and high operating pressure a priori are not available.

The main goal of this experimental work was the investigation of the response of non-premixed swirling flames to acoustic perturbations at various frequencies (0-350Hz) and the impact of the imposed air inlet velocity oscillations on soot formation and oxidation processes. These oscillations were realized with four loudspeakers. A natural gas/ethylene mixture oxidized by air under rich conditions (Φ = 1,56 and 42% C_2H_4 at P = 17,6 kW) is used in the experiments.

Soot particle sizing and soot detection measurements in highly turbulent flames at imposed air inlet velocity oscillations are performed using 2-Colour-TIRE-LII (2-Colour-Time-Resolved-Laser-Induced Incandescence). This method is a non-intrusive diagnostic technique based on the simultaneous detection of the time resolved LII signal at two different wavelengths. These measurements are combined with the imaging of OH*-Chemoluminescence and measurements of the temperature field. The presented experimental setup also allows the determination of normalized soot volume fractions. In order to avoid possible contributions from a fluorescence background ascribed to polycyclic aromatic hydrocarbons (PAHs) to the LII-signals, the LII-data was collected 39ns after LII_{max}-Signal. The reason is the much shorter relaxation time of the PAH fluorescence compared to the soot incandescence.

The experimental results give insights into the soot formation and oxidation processes and their interaction with the acoustic oscillations. A frequency depending decrease of the soot volume fraction, soot particle size and number density was observed. An explicit frequency dependence was found for the soot properties and sooting tendencies in this swirled turbulent diffusion flame due to changes in the mixing conditions between fuel and oxidizer, smoother temperature gradients and enlargement of the zone with high OH*-concentration (intensification of the soot oxidation). An air ratio fluctuations caused by the inlet velocity oscillations can be also a possible source of changes in the sooting conditions in the flame.

Keywords: laser diagnostics, turbulent flames, pollutant formation, flame instabilities

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Correlated laser-induced fluorescence of PAH and laser-induced incandescence to visualize soot inception in turbulent flames

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Laser-induced incandescence (LII) is a frequently used tool to monitor soot volume fraction distributions in academic and technical flames. Its application allows insight into soot formation and oxidation processes. It is widely accepted that LII is susceptible to mature soot while (mainly gaseous) organic precursor material will not be sufficiently heated up by comparable laser fluences to create significant LII emissions. In soot research, the process of soot inception is one of the least understood sub-mechanisms. Coincidently, the species contributing to this transition are not easy to access non-intrusively at flame conditions[1]. In LII research, one open question is the lower size limit of primary particles limiting applicability of this diagnostic tool or their internal structure and composition.

We applied simultaneous laser-induced fluorescence of PAH and laser-induced incandescence to sooting pressurized swirl flames. Excitation at 283 nm and detection in the spectral range between 300 and 400 nm yielded qualitative distributions of two to four-ring aromatics while also exciting LII emission of soot. Simultaneous irradiation with an IR laser sheet at 1064 nm excited only the soot without causing fluorescence of the aromatics. Comparison of the results from each technique therefore allows identification of PAH clouds. These appear as localized and isolated structures, more similar to soot than to OH signatures, and can either be linked to mature soot as evidenced by strong LII signal or as isolated events (see Fig. 1). Those PAH clouds linked to soot are indicative for soot inception. In addition, PAH clouds can be overlapped with a very faint LII signal only moderately stronger than image intensifier noise levels. In these cases LII seems to be a representation of nascent soot. Unlike in laminar flames, where PAH grow continuously into soot, turbulence allows for spatial separation of different maturity levels of soot formation. This shall be a valuable source of further understanding of soot formation.

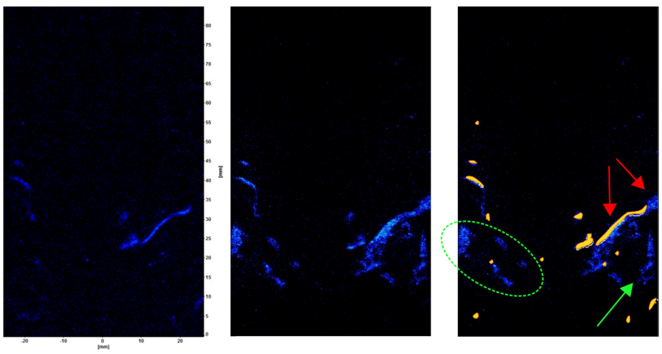

Fig. 1. Left image shows soot distribution (IR excitation), center plot is LIF/LII (UV excitation), right plot visualizes an overlay of the LII contour on the LIF/LII image. Separated PAH clouds are labeled green, transitioning ones into soot red.

Keywords: Soot inception, PAH LIF, LII, Turbulent flames

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Soot size and concentration in combusting sprays at high gas pressures and elevated temperatures estimated by optical methods

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A methodology for estimating soot particle size and concentration in turbulent combusting jets with elevated and inhomogeneous optical density is presented and discussed. The procedure is based on further developing the combination of quasi-simultaneous Laser Induced Incandescence (LII), Elastic Light Scattering (ELS) and Light Extinction (LE) measurements and exhibits a high potential for spatially resolved measurements of carbonaceous particles in flames and residual gases at a given instant. The method calculates the laser fluence across the flame and compensates for signal trapping, allowing measurements where laser extinction between the flame borders reaches values up to 90 %. The method was implemented by measuring particle size and concentration in the middle sagittal axis of optically dense, combusting diesel jets at a certain time after the start of combustion. Experiments were carried out in the Chalmers High Pressure, High Temperature (HP/HT) spray rig under conditions similar to those prevailing in direct injected compression ignition engines. Measurements of particle diameter, particle concentration and volume fraction conferring an instantaneous single-shot case and an average measurement from several consecutive sprays are presented and discussed.

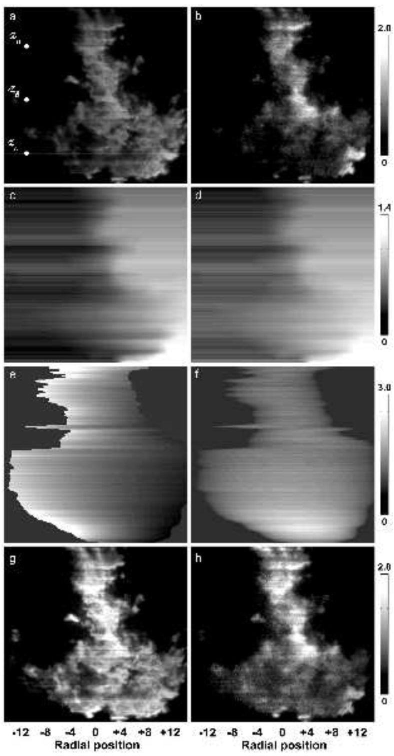

Fig. 1. a) Detected ELS image, b) Detected LII image, c) Calculated laser fluence for the ELS pulse, d) Calculated laser fluence for the LII pulse, e) Compensation matrix parallel to the laser plane (k_p), f) Line of sight compensation matrix (k), g) Compensated ELS image, d) Corrected LII image.

Keywords: Signal compensation, optically dense flames, combusting jets.

Laser-induced incandescence (LII) measurements on gas-borne silicon nanoparticles

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In the gas-phase synthesis of nanoparticles, e.g., in plasma or combustion processes, *in situ* analytical tools are rarely available to investigate particle formation, particle size, and particle size distribution. It is known that soot particle sizes can be observed by time-resolved laser-induced incandescence (TiRe-LII). LII shows a large potential, especially for elemental nanomaterials with high boiling points. In this paper, silicon nanoparticles synthesized in a microwave plasma reactor were investigated with LII downstream of the plasma zone.

Measurements were done at a pressure of 120 mbar using 3 slm Ar, 0.65 slm H_2, and 0.05 slm SiH_4 as core flow. The flow is surrounded by a coflow of 8 slm Ar and 1.5 slm H_2 for the stabilization of the plasma. These conditions typically lead to spherical, single crystalline silicon nanoparticles in the size regime of a few ten nm[1].

LII measurements were performed using a Nd:YAG laser at 1064 nm to heat the silicon particles above ambient temperature. Incandescence was measured at 442 and 716 nm simultaneously and the particle temperature was determined via two-color pyrometry (Fig. 1 (left), black line) and fitted to a bi-exponential function (red line)[2].

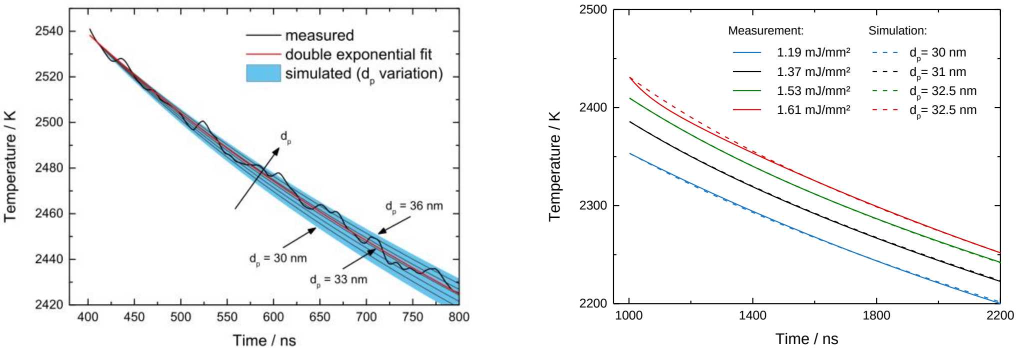

Figure 1. Measured and simulated temperature variation after laser heat-up for various particle diameters d_p (left); Comparison of measured (double exponential fit, solid lines) and simulated (dashed lines) signal traces for various laser fluences (right).

The time-resolved temperature profile was then used to determine the particle size of the silicon nanoparticles. Based on a heat transfer model taking into account nanoparticle cooling by evaporation, radiation, and heat conduction, a mean particle size around 30 nm could be calculated from the temperature decay. This is in almost perfect accordance with the diameters of particles scavenged from the gas flow determined from the specific surface area via nitrogen adsorption (BET) assuming monodisperse spheres. While increasing laser fluences results in increased particle temperatures, the calculated particle size is almost unaffected, cf. Fig. 1 (right).

Keywords: Silicon nanoparticles, laser-induced incandescence, particle size measurements, plasma synthesis

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Aerosol mass spectrometry of refractory black carbon containing particles

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Refractory black carbon (rBC) containing particles are formed in incomplete combustion processes with significant climate forcing impacts, due to strong light absorption, and potential health effects. The complex nature of these particles makes their characterization (microphysical, chemical and optical) challenging; new instruments are required for progress.

We have recently developed a new technique, the soot particle aerosol mass spectrometer (SP-AMS), to measure size-resolved mass and chemical composition of rBC particles[1]. The SP-AMS is equipped with an intracavity Nd:YAG CW laser vaporizer (1064 nm), similar to some laser induced incandescence (LII) techniques. Light-absorbing particles, such as rBC particles, absorb laser light and heat. Both the nonrefractory and refractory components of the particles vaporize. Particulate vapors are ionized by electron ionization (70 eV) and detected with high resolution time-of-flight mass spectrometry. Sample mass spectrum is shown in Figure 1.

We have participated in a number of recent laboratory studies and field campaigns focused on measuring laboratory flame soot[2], biomass burning, industrial flare emissions, internal combustion engine emissions[3,4], and urban environments[5]. As we continue to develop and apply the SP-AMS technique, we are focusing on (1) quantification; (2) characterizing carbon ion distributions generated by different rBC particle types; (3) identifying unique source-related chemical information for rBC-containing particles; and (4) characterizing the physical and chemical properties of these particles as a function of coatings due to gas-to-particle condensation (e.g. secondary aerosol material) and thermal denuding. An overview of the technique will be presented.

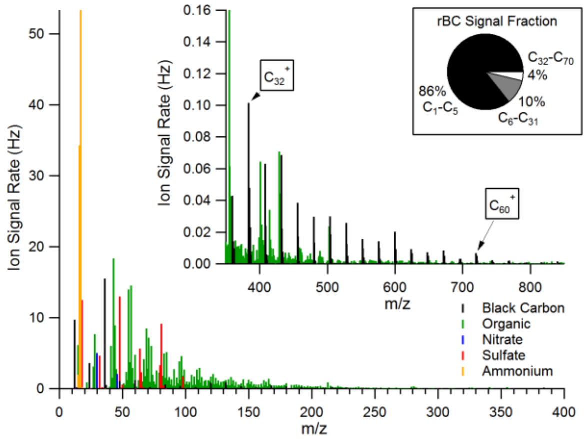

Figure 1. Refractory black carbon mass spectrum obtained from ambient aerosol sampled on Boston College campus.

Keywords: aerosol mass spectrometry, refractory black carbon, combustion soot.

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Abstracts

Posters

In situ analysis of the nanoparticle formation in the gas phase during carbon nanotube synthesis

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Temporally resolved laser-induced incandescence signals are from different gas phase particles during the growth of carbon nanotubes (CNTs) by floating catalyst chemical vapor deposition using the injection of aerosols containing both carbon feedstock (xylene) and catalyst precursor (ferrocene). The temporal decay of the LII signals allows quantitative measurements if the particle size which are in good agreement with electron microscopy observations. These results represent the first in situ analysis of the particle size in the gas phase during the CNT synthesis.

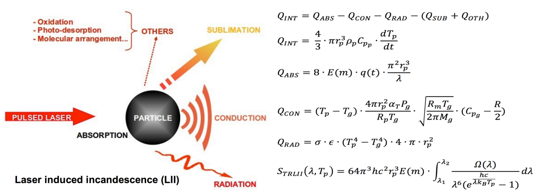

Fig. 1. LII theory and model, in which (Q_{SUB} + Q_{OTH}) sum can be neglected at low laser energy density (< 0.6J.cm^{-2})[1,2].

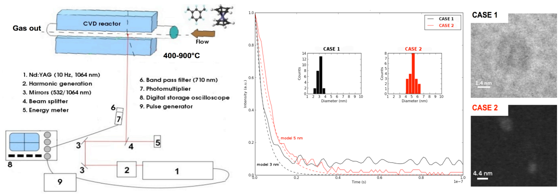

Fig. 2. LII diagnostic in a chemical vapor deposition reactor and comparisons between time-resolved microscopy.

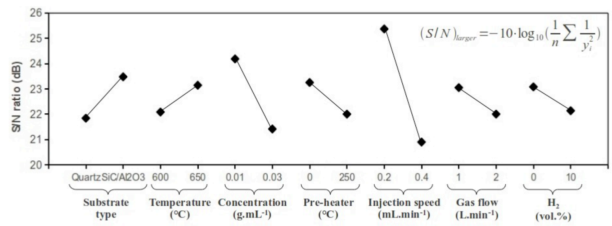

Fig. 3. Analysis of Taguchi robust design method introduced for multifactor process optimization[3]. A set of experiments was designed to investigate the most influential CVD parameters in the cluster size. Seven control factors with two levels were examined, leading to eight experiments organized in a L8 Taguchi matrix design.

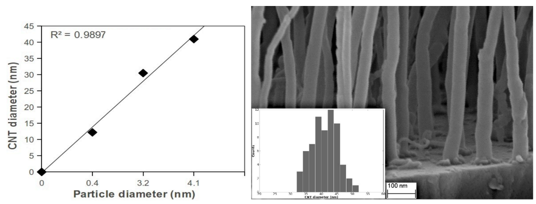

Fig. 4. Linear correlation between the particle size in situ detected by LII and the ex situ measured carbon nanotube (CNT) diameter. There is a direct relationship between the particle formed in the gas phase and the resulting nanotubes. These results present a first step toward the direct control of the CNT structure (i.e. diameter and chirality) during the growth process.

Keywords: LII, In Situ Analysis, Chemical Vapor Deposition, Taguchi Method, Carbon Nanotubes

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Mo nanoparticle sizing by Ti-Re LII and TEM

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The first attempt to extend LII diagnostic on Mo nanoparticles synthesized at photolysis of $Mo(CO)_6$ was undertaken by Murakami with co-authors[1]. However the independent particle size measurements were not performed and LII nanoparticle sizing results (600-3000 nm) were doubtful. Here we discuss the Mo nanoparticle sizing results obtained by Ti-Re LII and TEM. Mo nanoparticles were synthesized using multipulse excimer Kr-F laser photolysis of $Mo(CO)_6$ in a 0.5 cm^3 quartz reactor filled with an inert gas at room temperature. The pulse Nd:Yag laser (1064 nm) and two fast photomultipliers was used for two-color Ti-Re LII measurements at the wavelengths 488 (550, 400) and 770 nm. The Ti-Re LII measurements were based on the model described in[2], bulk Mo temperature dependent properties, and using the accommodation coefficient α=0.15 for Ar / Mo, recommended by Daun et al[3]. The mean Mo nanoparticle sizes measured by Ti-Re LII showed a gradual growth with number of photolysis pulses from 3 up to 14 nm (see Fig. 1) at the energy of Kr-F laser 20-30 mJ per pulse. The results of TEM sizing of nanoparticles synthesized at 6, 20, 30 and 600 pulses of Kr-F laser presented in Fig. 1 showed the higher mean values of nanoparticle sizes as compared with Ti-Re LII measurements. This discrepancy could be caused by the difference of the nanoparticle properties used in LII model from the bulk molybdenum. The dependence of peak temperature of Mo nanoparticles, synthesized at 20 Kr-F laser pulses, on YAG laser fluence presented in Fig. 2, could be an evidence of deviation of nanoparticle properties from the bulk ones. It is seen that the peak particle temperature does not exceed boiling temperature of balk Mo (4800 K) even at high fluences, when the Mo nanoparticle sublimation should be the case. On the other hand, to fit the LII and TEM data the thermal energy accommodation coefficient should be increased from 0.15 up to 0.3. Another assumption is that the volume of Mo nanoparticles increases owing to oxidation when particles are exposed in air during the time between their extraction from the reactor and TEM analysis.

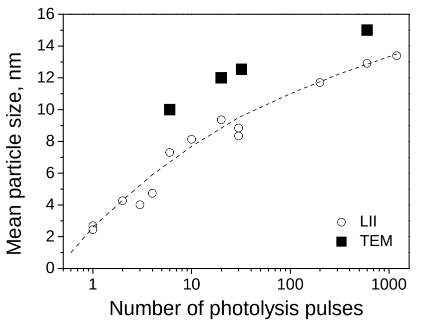

Fig. 1. The mean particle sizes measured by LII and TEM versus number of Kr-F laser pulses.

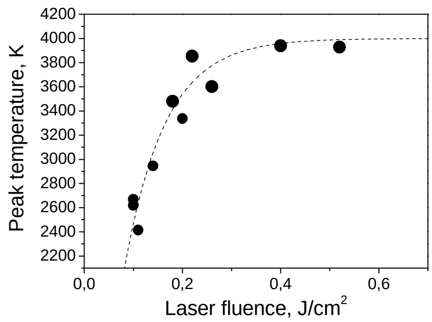

Fig. 2. The dependence of peak temperature of laser heated Mo nanoparticles on laser fluence.

This work has been supported by the RFBR (grant N 14-08-00505).
Keywords: Mo nanoparticles, Ti-Re LII and TEM sizing

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Soot measurements in premixed high-pressure flames using light emission, TiRe-LII, laser extinction, and TEM-sampling

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The quasi-simultaneous application of various soot characterization techniques in high-pressure flames such as light extinction, natural emission (in case of hot soot), laser-induced incandescence (LII)[1] and transmission electron microscopy (TEM) of deposited soot deposited on sampling grids it can be optically analyzed using either is challenging due to the complexity of sampling and providing optical access to such harsh conditions. We maintain a high-pressure burner facility that is capable to accomplish these tasks[2]. The burner stabilizes a rich premixed ethylene/air flame on a porous sintered stainless-steel plate which is surrounded by a rich methane/air flame and an air coflow to reduce heat loss and stabilizes the flame. Using time-resolved (TiRe-) LII for measuring soot particle size and volume fraction in high-pressure flames still is not well developed, which therefore is the main issue in our current work, also by validating results by independent calibration and sampling techniques.

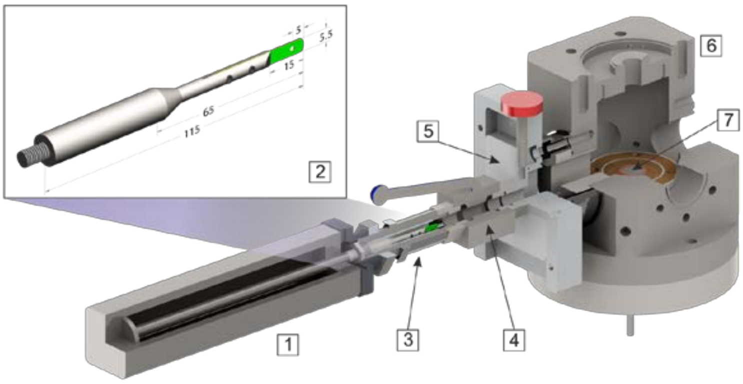

Fig. 1. High-pressure burner with attached TEM-sampler.

In the present work sooting premixed high-pressure ethylene/air flames at pressures from 1 to 25 bar are characterized by two-color TiRe-LII and laser-extinction measurements. Two-color TiRe-LII allows the determination of soot particle size, while the maximum intensity of the LII signal is assumed proportional to the soot volume fraction. In our experiments the heating laser is a Nd:YAG laser at (1064 nm) and the incandescence is detected at 550 and 694 nm, respectively by fast photomultipliers. The gas temperature was determined from fitting grey-body radiation spectra to the measured spectra. Laser extinction at 785 nm is measured to determine the soot volume fraction for comparison with the LII-derived values – especially taking into account the reabsorption of the LII signal at elevated pressures. A sampling unit has been developed to extract soot samples at various heights above the burner during high-pressure operation for *ex situ* TEM analysis. This additional information allows to improve the LII data evaluation and to improve heat transfer models suitable for high pressure environments.

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[1] C. Schulz, B. F. Kock, M. Hofmann, H. Michelsen, S. Will, B. Bougie, R. Suntz, G. Smallwood, *Appl. Phys. B* **83**, 333–354 (2006).
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LII in an Aero-Engine Exhaust Using a Low Peak Power Fibre Laser

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Keywords: Long-pulse LII, fibre laser, aero-engine

A commercial fibre laser (SPI G3 Laser) delivering 192 ns long pulses at a rep rate of 30 KHz has been used to observe LII in the exhaust of a modified helicopter engine. Images of LII, collected at 90°, from a focused Gaussian beam were recorded at average laser powers 2.2, 11 and 21 W. A schematic of the experimental arrangement is shown in Fig. 1.

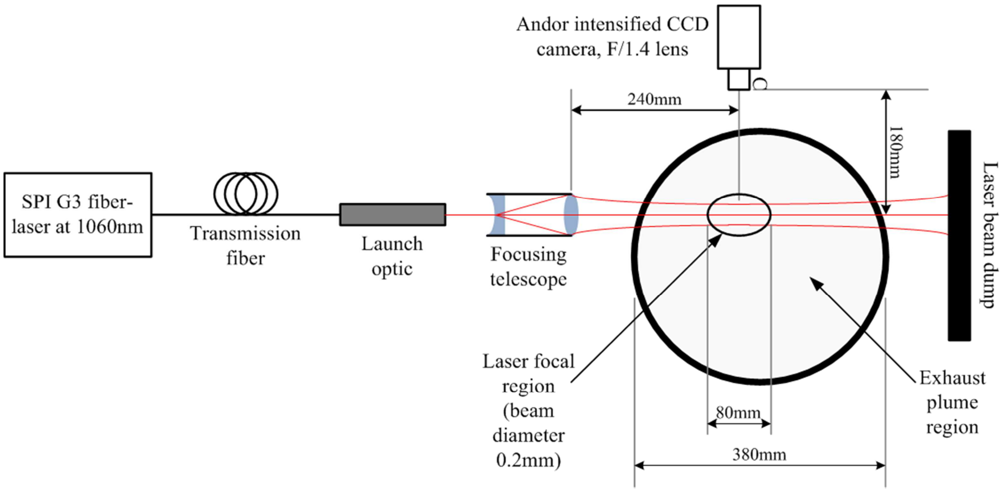

Fig. 1. LII arrangement for experiments on a modified helicopter engine

The waist diameter of the focused beam was 0.15 mm, corresponding to fluencies of 0.13, 0.64 and 1.21 J cm^{-2} for each power condition. As power increases, the distance along the beam over which LII can be detected increases and at 21 W a dip in LII intensity, expected due to particle vaporization at high fluence, was observed. By examining the signal level at different distances from the focus, fluence curves were constructed and compared with predictions[1]. During engine experiments gating of the camera was not possible and the minimum time integrated images were obtained over 0.01 s (300 pulses). The temporal behavior of LII using the SPI laser with a particle generator will be discussed.

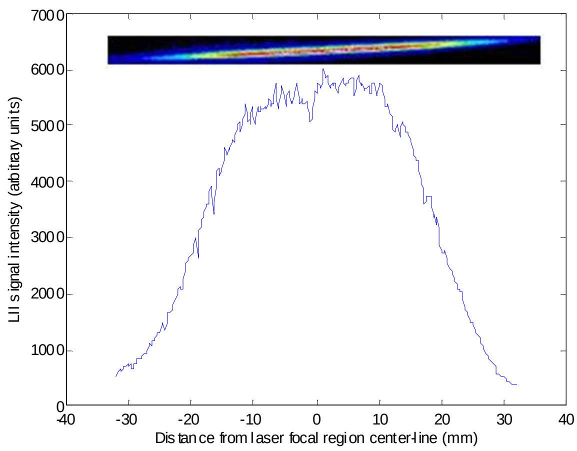

Fig. 2. An example of a 1s (30,000 laser pulse) image at 11W power

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Spectrally- and temporally-resolved laser-induced incandescence (LII) on gas-borne silicon nanoparticles with varying laser fluence

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Laser-induced incandescence (LII) is well-established for soot particle-size and volume-fraction measurements. The advantage against other laser-based measurement techniques is a relatively simple optical setup with high spatial and temporal resolution. With non-soot materials, (e.g. TiO_2)[1] it has been shown, that with increasing laser fluence a transition from LII to laser-induced breakdown emission (LIBS) occurs. In the present work a spectrometer/streak-camera combination is used to observe the temporal decay of the LII spectral emission after laser heating of gas-borne silicon nanoparticles. Measurements are carried out with varying laser fluence. Once the LIBS regime[2] is reached the involved species are identified.

Measurements are performed in a microwave-plasma nanoparticle-synthesis reactor approximately 30 cm above the nozzle at a pressure of 100 mbar. The initial gas flow consists of $SiH_4/Ar/H_2$ with 0.5/2.5/0.7 slm (standard liter per minute), surrounded by an Ar/H_2 sheath-flow with 7.5/1.5 slm for the stabilization of the particle-laden central jet[3].

A Nd:YAG laser at 1064 nm is used to heat up the silicon nanoparticles. The laser-induced emission was measured with a streak camera (Hamamatsu) attached to an imaging spectrograph. This system collected the emitted light in the 515–785 nm range over a time span of 2 µs with a temporal resolution of 3.9 ns. The recorded image is corrected for spatial sensitivity variation with a calibrated Ulbricht sphere (Fig. 1a). The maximum heat-up temperature as starting point for our heat transfer model is calculated by fitting Planck’s law to the mean value of 5 image rows around the highest intensity region of the recorded image (Fig. 1b). Figure 1c shows the resulting temperature decay curve including the simulation for a mean particle diameter of 27 nm.

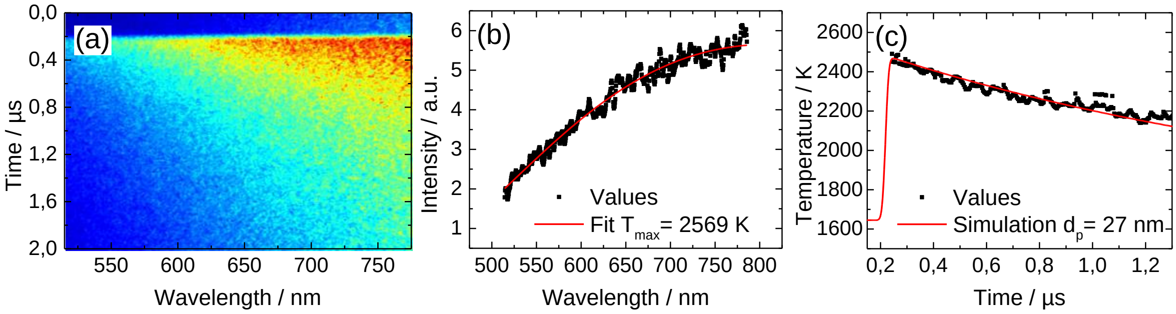

Fig. 1 (a) LII spectral decay. The maximum heat up temperature is fitted via Planck’s Law (b). The resulting temperature decay curve is shown in (c). *Squares:* Temperature decay fitted with Planck’s law for each time step. *Solid line:* Simulation of the temperature variation based on our heat transfer model.

No parasitic atomic line emissions were observed using moderate laser fluences of about 1.5 mJ/mm^2. The calculated particle diameter matches BET and TEM measurements very well. The heat-up temperatures determined from fitting the entire spectra yields lower temperatures than two-color thermometry (442 nm, 716 nm) for the same laser fluence.

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Quantitative Measurements of Soot Volume Fraction Using Planar LII in Diesel Spray Combustion under Diesel Engine Conditions

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Soot is one of the main pollutant emissions of diesel engines. Meeting the increasingly stringent emission regulations is the main motivation to study soot the formation mechanisms of soot during the diesel engine combustion processes and, ultimately, to improve the engine design to reduce soot emissions. The Laser Induced Incandescence (LII) technique has been proved to be a very effective method to measure the real-time soot concentrations. Although LII has been commonly used in soot measurements in laminar flames with success, challenges remain when it is applied to soot measurement in in-cylinder combustion and further studies are required.

The formation of soot during the combustion in diesel engines is strongly affected by the spray processes. The background pressure and temperature and the injection pressure also have a significant impact on the formation of soot. In this work, a high temperature and high pressure constant volume combustion vessel was used to simulate the in-cylinder temperature and pressure of top dead center of a diesel engine. The chamber was filled with high-pressure air and heating wire was used to heat the air.

A laser sheet of about 50 mm high and 1.5 mm wide at 532 nm and 10 Hz was used as the excitation source for planar LII imaging of soot distributions in the diesel spray at several moments after ignition. To quantify the soot volume fraction a two-color LII detection system (400 and 842 nm) was used to obtain the soot volume fraction at a selected location in the diesel spray. The two-color LII detection system was calibrated using a calibrated integrating sphere.

Keywords: Laser Induced Incandescence; Diesel Combustion; Soot Concentration

Application of Planar Laser Induced Incandescence in Turbulent and Sooting Flames: The Influence of Radiation Trapping and Beam Steering

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Laser-induced incandescence (LII) has become the leading technique for the measurement of soot in flames. Soot volume fraction (f_v), soot particle size (d_p) and the number (N_p) of primary soot particles can be measured and quantified using LII. The spatial distribution of these parameters can be achieved through planar laser-induced incandescence (PLII) which provides insight into soot gradients, sheet formation and interaction in practical turbulent flames and in engines.

Planar measurements of soot volume fraction in highly turbulent, non-premixed flames provide the key parameter to understand the interaction between soot chemistry and turbulence. However, quantitative measurement of soot volume fraction is still challenging due to several unresolved effects, e.g., beam attenuation, beam steering/dispersion and LII signal trapping especially in large flames. In the present work, the influences of LII signal trapping and beam dispersion on the results of f_v, performed in turbulent sooty flames are investigated, with the aim to estimate the experimental uncertainty and to propose a correction method.

The incandescent radiation, emitted from the laser heated soot particles is attenuated before reaching the detector resulting in an underestimation on f_v. This is not only because of the scattering and absorption from soot particles but also due to the absorption of polycyclic aromatic hydrocarbons (PAH) present in the optical path between the measurement volume and the detector. Typically, the incandescence in the wavelength range of 400 – 450 nm is collected as the LII signals. Hence, at these short wavelengths, the signals are more prone to attenuation than are those at the longer wavelength. While signal trapping can be estimated theoretically when quantifying the LII signal attenuation from soot particles only is considered leading to an underestimation of this effect.

In addition, the laser beam dispersion, due to the strong thermal gradation in turbulent flames, is another potentially uncertainty in quantifying the LII measurements. The dispersion will result in an extension to the laser beam sheet thickness. This in turn will not only result in a lower spatial resolution but also influence the recorded LII signals. The laser fluence is reduced by soot scattering/absorption. To overcome the influence of this attenuation, an infrared laser beam (1064 nm) with a saturated fluence is typically used to significantly reduce the absorption/scattering and to ensure the LII signal is independent to the laser fluence. However, as the collected LII signal is linearly proportional to the measurement volume, it will be affected as a result of the change in the laser beam thickness.

The influence of the signal trapping on the LII measurements in a turbulent non-premixed C_2H_4/air flame will be presented. It has been studied by recording the transmitted intensity of a 450 nm laser beam after passing through flame. The effect of dispersion is systematically studied by recording the transmitted profile of the LII laser beam through a set of different turbulent C_2H_4/air flames.

Real-time Capable Characterization of Soot Nanoparticles by Wide-Angle Light Scattering (WALS)

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For the industrial production of nanoparticles a non-invasive online-characterization technique is desired to control the production-process. Elastic light scattering (ELS) allows for the determination of aggregate sizes and fractal dimensions of particle aggregates[1], yet conventional approaches suffer from either slow measurement rates or limited information when scanning goniometers or few detectors are used simultaneously.

Oltmann et al.[2,3] developed the wide-angle light scattering (WALS) method to determine aggregate sizes in flames. In this approach an ellipsoidal mirror images the light scattered from the measurement volume located in one focal point of the mirror into an aperture at the second focal point and onto a CCD camera chip. With this approach a scattering diagram with an angular range from 10° to 170° and a high angular resolution of 1° and below can be obtained from single-shot images. A calibration image accounts for optical properties of the set-up.

For industrial applications a mobile demonstrator employing a compact laser system (18 mJ/pulse @ 532 nm, 100 Hz) and camera (max. 100 Hz @ 500 x 500 pixels, 8-16 bit) was developed, which also comprises a smaller mirror with $\Delta f = 360$ mm. As data acquisition and evaluation have to be performed online, fast and robust algorithms have been developed. The scattering data is evaluated within 100 ms and a feedback about the radius of gyration is given to the user. For obtaining additional information on particle properties the WALS-approach is favorably combined with extinction or laser-induced incandescence (LII)[4] measurements.
Measurements were carried out on soot particles from a flat-flame burner (McKenna type). Results show good agreement with TEM data.

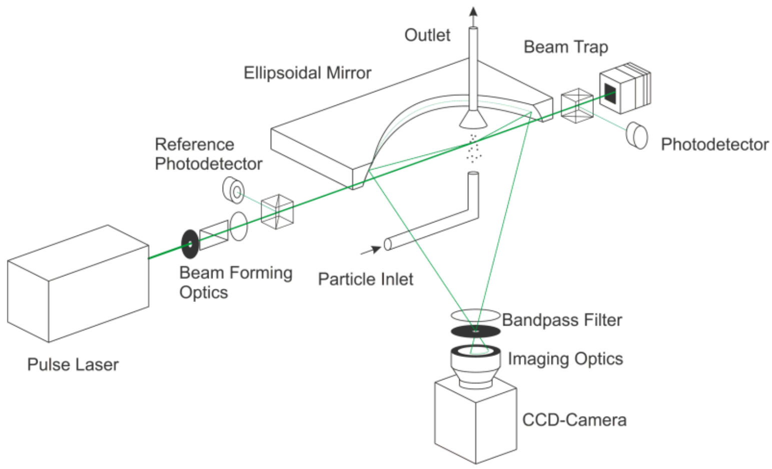

Fig. 1. Sketch of experimental WALS setup.

Keywords: elastic light scattering, laser-induced incandescence, nanoparticles

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4 C. Schulz, B.F. Kock, M. Hofmann, H. Michelsen, S. Will, B. Bougie, R. Suntz, G. Smallwood, *Applied Physics B* **83**, 333-354 (2006)

Approach to standardize a spray-flame nanoparticle synthesis burner

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Spray-flame synthesis is frequently used for generating metal oxide and ceramic nanoparticles. A few concepts are frequently used in several labs and are commercially available[1], but they are not well suited for fundamental studies because their geometry is neither ideal for *in situ* measurements such as particle-sizing with LII nor easy to simulate. For laminar sooting flames, well characterized standard flames[2] have been developed and used for modeling soot chemistry. Also for non-sooting turbulent flames, standardization has been used for a long time to advance both measurement technology and simulation through direct interaction[3]. This work focuses on the development and characterization of a standardized burner for spray-flame nanoparticle synthesis.

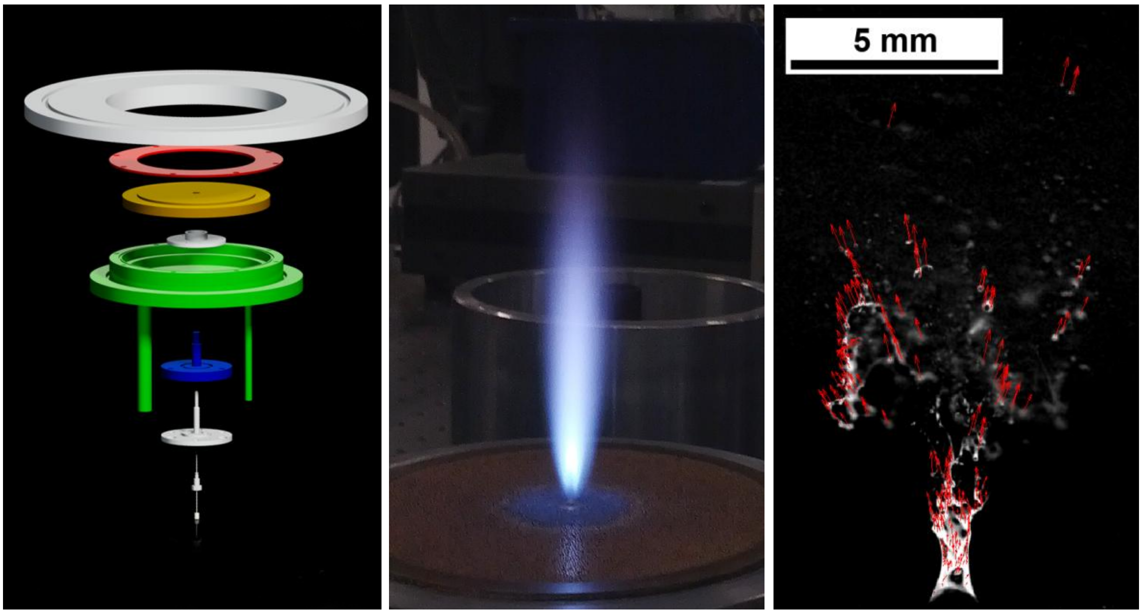

Fig. 1: Left: Assembly drawing of the nozzle of the spray burner, center: Ethanol spray flame, right: Particle tracking velocimetry to determine droplet velocity.

Many precursors for nanoparticle synthesis can be dissolved in alcohols or aromats (e.g., ethanol and toluene). Therefore, the nanoparticle forming precursor can be introduced into the flame together with the liquid fuel. The transition position between pilot-flame and co-flow is designed to be blurred to enable computational fluid dynamics simulations with moderate mesh resolution in regions that are not relevant for the process of interest. Particle-tracking velocimetry and NO-LIF thermometry were applied to the new burner to investigate droplet-velocity and temperature fields. This burner can be operated with up to 500 slm (standard liter per minute) compressed air or nitrogen as co-flow for effective temperature quenching. The operating pressure range reaches from 75 mbar up to several bar. Figure 1 shows a side view of the novel spray-flame nanoparticle-synthesis burner operated with ethanol.

Keywords: flame-spray nanoparticle-synthesis, computational fluid dynamics

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Soot optical properties investigation by two-color laser-induced incandescence measurements

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The need to measure, characterize and monitor carbonaceous particles emission triggered the interest for the development of advanced diagnostic techniques based either on their thermal or optical properties. Laser-Induced incandescence technique (LII) has been proved to be a powerful tool able to measure concentration and size of carbonaceous particles. Aim of this work is to further investigate the capability of the two-color LII technique to be applied for environmental applications such as ambient air quality and source emission monitoring. Moreover, since LII signals strongly depend on different parameters, such as optical and heat-exchange properties of the particles as well as the laser density energy[1] , the technique has been tested as a tool to investigate the optical properties of carbonaceous particles under different experimental conditions. In fact, by using different laser density energies, the particle absorption properties, and consequently the nature of the detected particles, can be investigated.

Measurements of carbonaceous particles were performed in different environmental conditions, covering a wide range of concentration (from ambient air to cars' exhaust), varying the laser density energy. The measurements were carried out with a high sensitivity LII portable instrument recently developed in our lab[2].

The results show a significant difference in the incandescence ratio behavior at low laser density energies, suggesting a difference in the absorption properties and in particular in the refractive index of carbonaceous particles under investigation.

Keywords: two-color LII, soot optical properties, particulate emission

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Determination of small soot particles in the presence of large ones from time-resolved laser-induced incandescence

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Information about the polydispersity of soot can in principle be gained from time-resolved laser-induced incandescence (LII) using pre-assumed particle-size distributions. This paper introduces an alternative method, designated two-exponential reverse fitting (TERF). It is based on mono-exponential fits to the LII signal decay at a delayed time. The method approximates the particle-size distribution as a combination of one large and one small monodisperse equivalent mean particle size and does not require a distribution assumption. It also provides a ratio of the contribution of both size classes. The systematic error caused by describing LII signals by mono-exponential decays is calculated as less than 2% for LII signals simulated for monodisperse aggregated soot with heat-up temperatures for which evaporation is negligible. The effects of particle size, heat-up temperature, aggregate size and pressure on this error are evaluated. The method is tested on simulated LII signals for lognormal and bimodal size distributions and applied to LII data acquired in a laminar non-premixed ethylene/air flame at various heights above burner. The results are compared to transmission electron microscopy (TEM) measurements of thermophoretically-sampled soot. The particle size of the large particle-size class evaluated with the method showed good consistency with TEM results, however the size of the small particle-size class and the relative contribution could not be compared due to missing information in the TEM results for small particles. These limitations of TEM measurements is discussed and the effect of the exposure time of the sampling grid is evaluated.

Keywords: TiRe LII, particle-size distribution, TERF method, TEM

Sensitivity analysis for in-cylinder soot-particle size imaging with laser-induced incandescence

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Soot particle sizes can be determined from time-resolved laser-induced incandescence (LII) in point measurements where full signal traces are detected. For instantaneous imaging, strategies are required that must cope with time-gated information and that rely on assumptions on the local boundary conditions. A model-based analysis is performed to identify the dependence of LII particle-size imaging on the assumed boundary conditions such as bath gas temperature, pressure, particle heat-up temperature, accommodation coefficients, and soot morphology. Various laser-fluence regimes and gas pressures are considered. For 60 bar, fluences that lead to particle heat-up temperatures of 3400–3900 K provided the lowest sensitivity on particle-sizing. Effects of laser attenuation are evaluated. A combination of one detection gate starting at the signal peak and the other starting with 5 ns delay was found to provide the highest sensitivity at 60 bar. The optimum gate delays for different pressures are shown. The effects of timing jitter and poly-dispersity are investigated. Systematic errors in pyrometry imaging at 60 bar is evaluated.

Keywords: Soot particle-size imaging, high pressure, sensitivity analysis

Assessment of soot particle-size imaging with LII at Diesel engine conditions

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Two-time-step laser-induced incandescence (LII) imaging was performed in Diesel engine-relevant combustion to investigate its applicability for spatially-resolved measurements of soot primary particle sizes. The method is based on evaluating gated LII signals acquired with two cameras consecutively after the laser pulse and using LII modeling to deduce particle-size from the ratio of local signals. Based on a theoretical analysis, optimized detection times and durations were chosen to minimize measurement uncertainties. Experiments were conducted in a high-temperature high-pressure constant-volume pre-combustion vessel under the Engine Combustion Network's (ECN) "Spray A" conditions at 61–68 bar with additional parametric variations of injection pressure, gas temperature and composition. The LII measurements were supported by pyrometric imaging measurements of particle heat-up temperatures. The results were compared to particle-size and size-dispersion measurements from transmission electron microscopy (TEM) of soot thermophoretically sampled at multiple axial distances from the injector. The discrepancies between the two measurement techniques are discussed to analyze uncertainties and related error sources of the two diagnostics. It is found that in such environment where particles are small and pressure is high, LII signal decay times are such that LII suffers from a strong bias towards large particles.

Keywords: Soot particle-size imaging, Diesel jet, ECN, TEM, pyrometry imaging

Soot volume fraction measurement by extinction and Laser Induced Incandescence in a wood-fired boiler under varying boiler conditions.

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Biomass represents approximately 14% of the worldwide energy consumption. Unfortunately, its combustion generates a number of air pollutants including particle emissions containing both large fly ashes (>1 µm) and fine soot particles (<1 µm). This study aims to investigate a typical wood pellet-fired boiler dedicated to domestic heating with a nominal thermal capacity of 30 kW. Soot volume fraction and paticle size distribution have been measured in the exhaust of the boiler. In-situ and ex-situ techniques are investigated to this aim and measurements are performed during the slumber and the steady-state mode for different working conditions of the boiler (Full and 50 % loads). For in-situ measurements, a chimney with three optical accesses is implemented to the exhaust gas pipeline. Soot volume fraction is measured using the two-color LII technique (2C-LII): LII signal excited at 1064 nm is detected at 530 and 700 nm. The detection efficiency of the laser system has been determined by using a calibrated integrating sphere, leading to an instantaneous measurement of the absolute soot volume fraction. In parallel, extinction technique is applied to measure the volume fraction of the nano and micro sized absorbing particles (soot and ashes). Measurements are performed with a Continous Wave laser at 1064 nm. The incident and transmitted beams are recorded simultaneously with the LII decay time of soot excited with the pulsed Nd-YAG laser. The comparison between both techniques allows the determination of soot concentration (with 2C-LII) and the total particle concentration (ash and soot) with extinction.

In parallel, the coupling of these two techniques was validated in a laboratory sooting flat flame of ethylene stabilized on a porous burner.

Based on E(m) evolution versus wavelength[1], the auto-absorption of LII photons have been calculated in order to correct the soot volume fraction measurement. Finally, the size distribution of particles is measured using a SMPS (scanning mobility particle sizer) in the combustion chamber (brazier) and in the exhaust gas. The scattering coefficient has been evaluated in order to correct the absorption coefficient initially measured by extinction[2].

The profiles of the particle concentrations versus time clearly show cyclic peaks related to the pellet feeding.

Keywords: absolute volume fraction, soot, LII, extinction, wood boiler

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Measurement of soot temperature, concentration and cooling rate; and bulk fluid temperature using modulated laser induced incandescence

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Modulated LII, where a chopped laser light source was employed to generate a modulated LII signal in a laminar diffusion flame was recently introduced[1]. The use of a modulated LII 2 colour pyrometry technique (MLII) to measure soot temperature in a laminar diffusion flame is described and the results compared to CARS gas temperature experiments. A sinusoidal modulated diode laser is used to excite the soot and both the modulated LII intensity and its relative phase to the excitation source are measured with a lock-in amplifier and recorded. Modulation frequencies from 25 Hz to 40,000 Hz were employed.

The temperatures derived from the ratio of the modulated LII radiation signal at 445 nm and 750 nm were largely independent of modulation frequency and compared well with bulk gas temperature obtained by CARS spectroscopy[2].

A theory is developed to explain the dependence of the phase delay of the modulated LII signal (with reference to that of the laser excitation source) on gas replacement time in the sample volume, soot cooling rate and soot volume fraction. The theory is shown to give a reasonable fit to the experimental results at all frequencies as shown in Figure 1. At lower frequencies the phase delay is dominated by the gas replacement time in the sample volume which can be related to the bulk fluid velocity and at higher frequencies by the cooling rate of the heated soot. Time constants for both processes and the soot volume fraction are derived from the data and shown to be largely in agreement with the expected values.

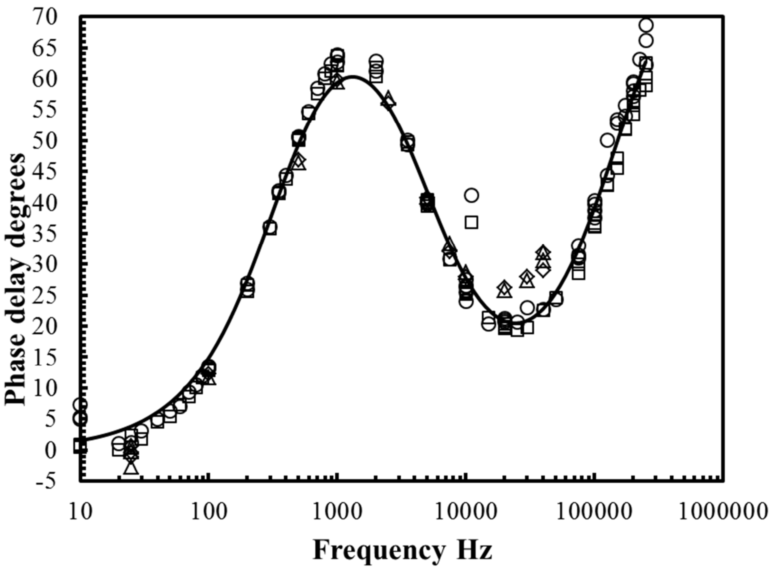

Fig. 1: Measured phase delay of MLII signals with respect to laser excitation source. Open squares, and open circles phase from 445 and 750 nm channels, respectively using 7265 lock-in amplifier. Open triangle, and open diamond phase from 445 and 750 nm channels, respectively using Femto lock-in amplifier. Curve least-mean-squares fit of data using developed theory.

Keywords: modulated LII, temperature, soot optical properties

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Probing the smallest soot particles in low-sooting premixed flames using laser-induced incandescence

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In this work we investigate nascent soot particles by analyzing laser-induced incandescence (LII) signals obtained in low-sooting premixed flames. The analysis covers two data sets obtained in separate experimental campaigns. The first data set was obtained in a previous work[1] in methane/oxygen/nitrogen flames (equivalence ratio range $1.95 < \Phi < 2.32$) stabilized at 26.6 kPa whereas the second was obtained in atmospheric ethylene/air flames ($1.77 < \Phi < 2.00$).

Both studies show similar trends, i.e. a gradual change of the fluence curves (evolution of the LII signal as function of the laser fluence) from the well-known S-shaped curve for mature soot found at high heights above the burner (HAB) and high equivalence ratio to a nearly linear behavior for nascent soot found at low HAB and reduced equivalence ratio (Fig. 1). With this change comes a decrease in the LII decay time (and hence inferred particle size) which appears to be almost constant with HAB in flames having the lowest equivalence ratio at which the incandescence signal could be detected. In these flames, so-called nucleation flames, the stability of the particle size with HAB suggests that recently nucleated particles have undergone very slow surface growth and coagulation. Experimental results are analyzed by using a theoretical model for LII to determine the particle diameter evolution throughout the flame at various experimental conditions. We highlight the size difference from nascent soot particles up to mature soot, giving insight into the particle nucleation and the surface growth processes as a function of reaction time and flame conditions.

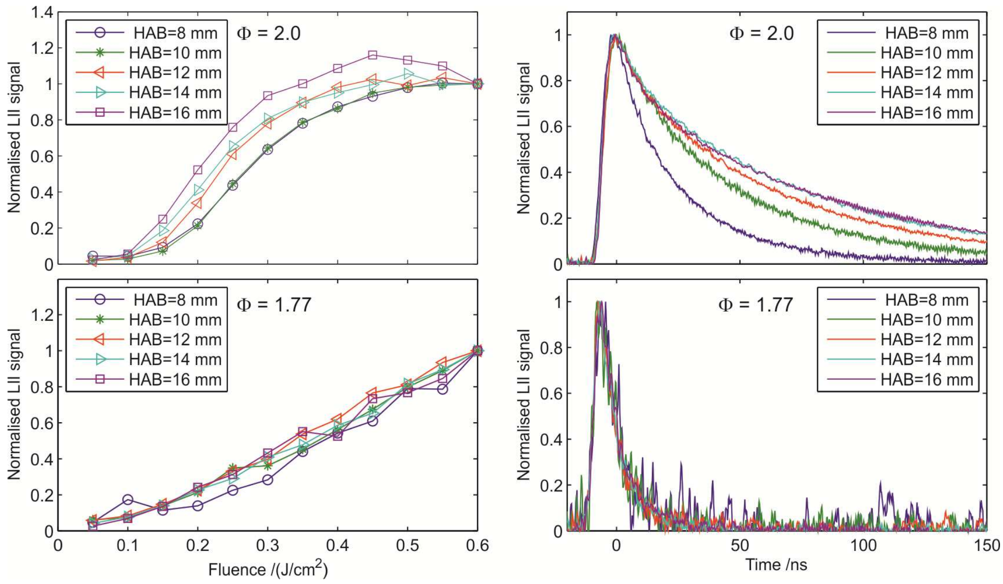

Fig. 1. Fluence curves and time-resolved LII signals (laser fluence 0.2 J/cm^2) for a moderately sooting flame (top) and a flame at the sooting threshold (bottom).

Keywords: Low-sooting premixed flames, Optical properties

[1] Mouton et al. Appl. Phys. B, vol.112 (2013) 369-379

Mini-CAST as a Cold Soot Source for Studying Optical Properties of Well Characterized Carbonaceous Particles

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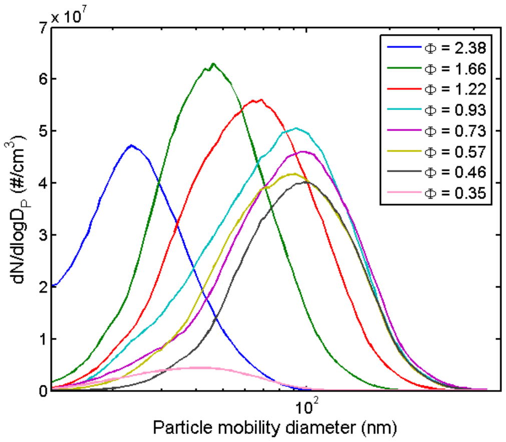
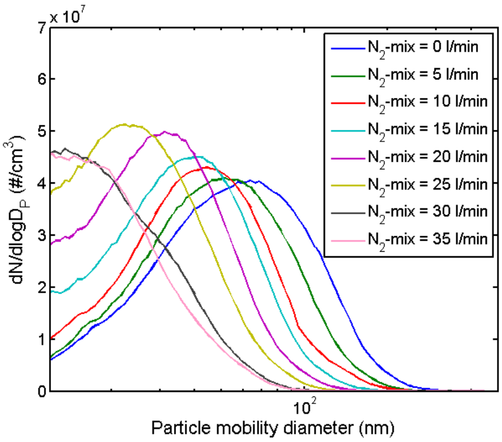
In the latest report from IPCC[1] (2013) aerosols are discussed as one of the most uncertain parameters in estimations of the radiative forcing, one of the most important aerosol being black carbon (soot). It is well known that soot particles leaving a combustion process, transforms both morphologically and chemically in the atmosphere. In order to improve the knowledge about soot formation and transformation, controlled laboratory experiments can be performed to measure various optical and structural properties. In this way the soot-radiation interaction can be better estimated and understood.

Carbonaceous particles in the size range of 10-150 nm of different chemical composition (black carbon BC/brown carbon BrC) are generated using a Mini-CAST (Combustion Aerosol Standard) model 6203C by quenching a diffusion propane/air flame of different equivalence ratios (Φ), and optionally introducing a N_2 mixing gas into the fuel flow. Initial studies have been made using a Single Particle Aerosol Mass Spectrometer (SP-AMS), a Scanning Mobility Particle Sizer (SMPS), and an Aethalometer. In Fig. 1 some results of the soot size distribution is presented for different Φ and in Fig. 2 for different N_2 mixing gas flows. In the following work we intend to use Laser Induced Incandescence (LII) along with Elastic Light Scattering (ELS) and extinction measurements, to study optical properties of soot and couple these to certain well characterized morphological and chemical features.

We investigate Mini-CAST (model 6203C) as a possible standard soot source within the LII community, and we aim to use this instrument to study soot particle transformation on a laboratory scale. Using a stable cold soot target source[2] with the ability to generate a wide variety of soot types, not only the different kinds of soot can be better characterized, but also different soot LII signals can be further investigated.



Figur 1. SMPS data presenting the particle size distribution for different equivalence ratios (Φ).



Figur 2. SMPS data presenting the particle size distribution as different N_2 mixing gas flows are introduced into the fuel flow.

Keywords: Soot, LII, ELS, extinction, Mini-CAST, SP-AMS, SMPS

1 IPCC, *WG I: The Physical Science Basis* (2013).
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Studies of optical and physical properties of soot in premixed flat flames using laser-induced incandescence, elastic light scattering and extinction

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The soot optical properties are dependent on the wavelength[1] and the maturity of the soot[2]. Studies of these properties are therefore of great importance, for example to get accurate soot volume fractions from optical diagnostic techniques. In this work two premixed ethylene/air flames (Φ = 2.1 and 2.3) on a bronze McKenna burner have been investigated using laser-induced incandescence (LII), elastic light scattering (ELS) and extinction.

Investigation of wavelength dependence of the soot properties in the visible and near infrared region (404 – 1064 nm) have been performed using extinction measurements with diode lasers (Fig. 1). Measurements show an increase of the inferred soot volume fraction with decreasing wavelength (assuming absorption described by RDG theory, a constant refractive index and neglected influence of scattering). The increased inferred soot volume fractions for wavelengths shorter than ~700 nm are assumed to be mainly caused by polycyclic aromatic hydrocarbons. Accurate soot volume fractions are important for calibration of techniques such as LII.

A pulse-probe experiment was conducted using two pulsed lasers, one at 1064 nm to heat the particles and to be used for LII and one at 532 nm to probe the scattering from the heated particles. The results indicate that more nascent soot particles are absorbing less laser light, i.e. are more transparent (Fig. 2), in good agreement with previous work by Olofsson et al.[3] Additionally 2-color peak temperatures for the two flames have been derived.

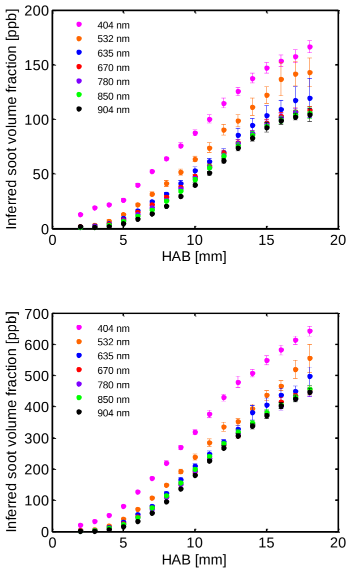

Fig. 1. Inferred soot volume fraction as function of height above burner (HAB) for various laser wavelengths for two flames, Φ = 2.1 (top graph) and 2.3. Evaluated with m = 1.56 - 0.46i[4]

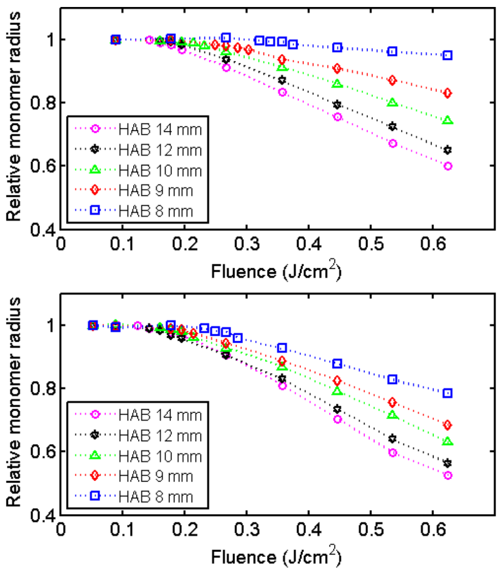

Fig 2. Inferred relative monomer radius from ELS as function of laser fluence at 1064 nm for two flames, Φ = 2.1 (top graph) and 2.3.

Keywords: LII, ELS, Extinction, Soot, Absorption function

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Effect of primary particle polydispdersity on the absorption cross section of soot aggregate and the implications to the soot absorption function derived from low-fluence LII

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The absorption and emission properties of soot aggregates are critical properties in laser-induced incandescence (LII) of soot. Under conditions of known laser pulse, namely wavelength, duration, and fluence, the soot aggregate absorption cross section controls the maximum temperature that soot aggregates can reach in low-fluence LII. This idea has been used in several previous studies to estimate the soot absorption function E(m) in both laminar diffusion and premixed flames. However, the theory used in those studies to obtain the soot aggregate absorption cross section was the Rayleigh-Debye-Gans approximation for fractal aggregates (RDG-FA) and the effect of primary particle polydispersity was neglected.

Previous results obtained using more accurate numerical techniques including the discrete dipole approximation (DDA) and generalized Mie-solution method (GMM) for soot aggregates formed by monodisperse primary particles showed that RDG in general underestimates soot aggregate absorption by about 10%, depending on the aggregate size and the laser wavelength, for typical aggregate sizes encountered in laminar flames. It implies that the derived E(m) value could be about 10% higher than the true E(m) value.

Primary soot particles within a given aggregate are not uniform in diameter but display a distribution with about 20% deviation from the mean. There has only been one study about the effect of primary particle polydispersity on the absorption and scattering properties of soot aggregates[1]. Farias et al. [1] proposed that the absorption cross section of a soot aggregate is enhanced by a factor of $1+3(\sigma_{dp}/d_{p,mean})^2$, independent of the aggregate size.

In this study the absorption cross section of soot aggregates formed by polydisperse primary particles was investigated by both the RDG-FA and GMM. Fractal aggregates formed by monodisperse and polydisperse primary particles were numerically generated by a diffusion-limited cluster aggregation (DLCA) algorithm. The polydisperse primary particles follow the Gaussian distribution with a typical 20% deviation. For soot aggregate sizes between 20 and 291 neglect of primary particle polydispersity leads to about 8 to 18% underestimation of soot aggregate absorption cross section, which is even more significant than the effect of multiple scattering and is considered significant in the context of LII. These results suggest that the E(m) values derived from low-fluence LII are more significantly overestimated than previously thought.

Keywords: Soot aggregates, absorption cross section, RDG-FA, polydisperse primary particle.

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Comparison of LII and Extinction Measurements of Soot Volume Fraction in Turbulent Jet Flames

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The quantification of even temporally averaged soot volume fraction and the statistics of its spatial and temporal distribution in turbulent jet flames is challenging, particularly in moderately sooting environments in which a significant fraction of the incident laser light and generated LII signal is absorbed and scattered. As part of a project designed to capture data for comparison with turbulent flame models of soot formation and radiation, we have performed both conventional (high fluence) LII imaging and localized laser extinction measurements within turbulent jet flames fueled by ethylene and a prevaporized JP-8 surrogate. The extinction measurements were performed using a narrow ceramic-tipped optical probe that defined a 10 mm long sample path within the investigated flames. The chosen extinction wavelength (635 nm) was similar to the LII detection wavelength (300 – 600 nm), thereby minimizing uncertainties related to the spectral dependence of soot optical properties when comparing results from the two techniques. In general, the extinction-based measurements would be expected to have less susceptibility to errors and needed corrections, because of its insensitivity to laser absorption and signal trapping effects. The extinction measurement suffers from poor spatial resolution, but this should not be a limiting factor when comparing temporally averaged signals in flame regions downstream of the initial soot formation zone. Comparison of the extinction signals with LII-derived soot f_v clearly shows corrections are needed for quantifying LII in the JP-8 surrogate flame. For the ethylene flame, with reduced soot concentrations, the needed corrections are much smaller. With suitable corrections to the LII signals, the mean soot volume fractions measured with both techniques are in reasonable agreement, lending credibility to the correction approaches taken and to the absolutely magnitude of soot volume fraction measured, within the accuracy of the assumed dimensionless soot extinction coefficient, based on measurements in laminar flames.

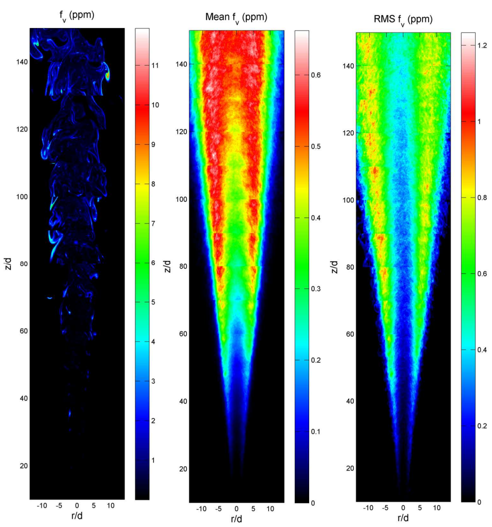

Fig. 1. Stacked LII planar images of instantaneous f_v, mean f_v (1000 images) and RMS of f_v in a turbulent jet flame fueled by prevaporized JP-8 surrogate

Keywords: soot, LII, extinction, volume fraction, absorption

Soot Measurements in Counterflow Non-premixed Flames using Laser Induced Incandescence: Soot Volume Fraction, Particle Size, and Number Density

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The counterflow burners provide a useful canonical configuration to numerically and experimentally analyze basic effects of soot nucleation, growth, and agglomeration under various conditions, namely fuel and oxidizer compositions, pressures, and residence times. Even in this simple reacting flow configuration, large spatial gradients of composition and temperature across the reaction layer present challenges to soot measurement techniques.

An absolute irradiance calibrated two-color time resolved Laser Induced Incandescence (LII) technique was utilized to collect quantitative soot incandescence data for determination of soot particle temperature, primary particle size, soot volume fraction, and number density. The approach requires a comprehensive analysis of the LII nano-scale heat transfer model. In this work, all the sources of uncertainty from both measured experimental variables and assumed heat transfer model variables were considered and perturbed with respect to their uncertainty bounds to obtain overall uncertainty of the measurements. The resulting overall uncertainties of soot volume fraction, primary particle size, and soot number density are reported here.

The absorption function was identified as the key source of uncertainty in determining soot volume fraction in counterflow flames. Uncertainty in the primary particle size was dominated by uncertainty in the thermal accommodation coefficient and ambient gas temperature. Since the experiment repeatability uncertainty was much lower than combined total uncertainty due to heat transfer model and other experimental variables, accurate heat transfer model parameters are needed for reducing the overall uncertainty of LII measurements.