FOURTH INTERNATIONAL WORKSHOP AND MEETING ON

LASER-INDUCED INCANDESCENCE: QUANTITATIVE INTERPRETATION, MODELING, APPLICATION

April 2010, Varenna, Lecco, Italy

Synopsis of recent LII research activities as presented on the 4th international workshop on LII

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Combustion Research NRC · CNRC Institute for Chemical Process and Environmental Technology

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Introduction

- International LII community first met in 2005
- Typically representing about 10 research groups world-wide
- Goal is a deeper understanding of the process of laser-induced incandescence
  - for more accurate determination of particle concentrations and sizes
    - inside and post-combustion
    - soot and non-soot
  - through regular intensive exchange of experience
  - for improved application to relevant / industrial processes
- Developing LII for technical application
- Has been key contributor to the IEA nanoparticulate diagnostics collaborative task

2010 LII Workshop Structure

- **Session 1 - Soot properties and LII signal in standard flames**
  - optical properties
- **Session 2 - LII models and validation**
  - cancelled due to limited air traffic
- **Session 3 - Extending LII**
  - combining LII with other techniques
  - comparing LII to other techniques such as SMPS, PIMS, fluorescence
  - nascent soot particles
- **Session 4 - LII under various conditions**
  - high/low pressure conditions
  - turbulent and technical flames, engines
  - environmental applications

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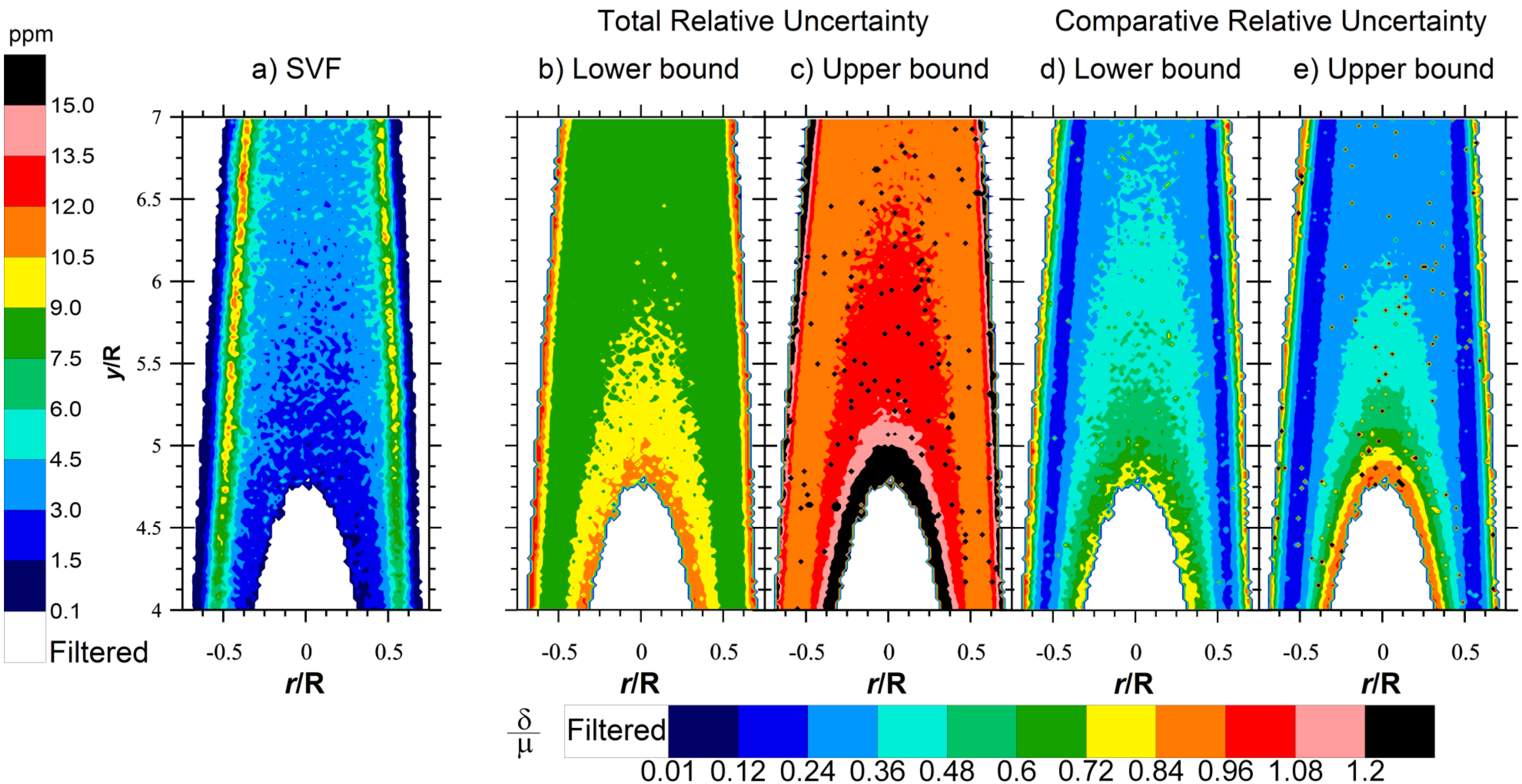
Session 1 - Soot properties and LII signal in standard flames

- We know/assume that laser-heated soot particles emit LII signal even if the laser pulse modifies the particles
- Question: how much are optical soot properties changed and how significant is the influence on the precision for particle sizes or soot concentrations?
- Different groups designed experiments approaching these issues

Monte-Carlo Uncertainty Results



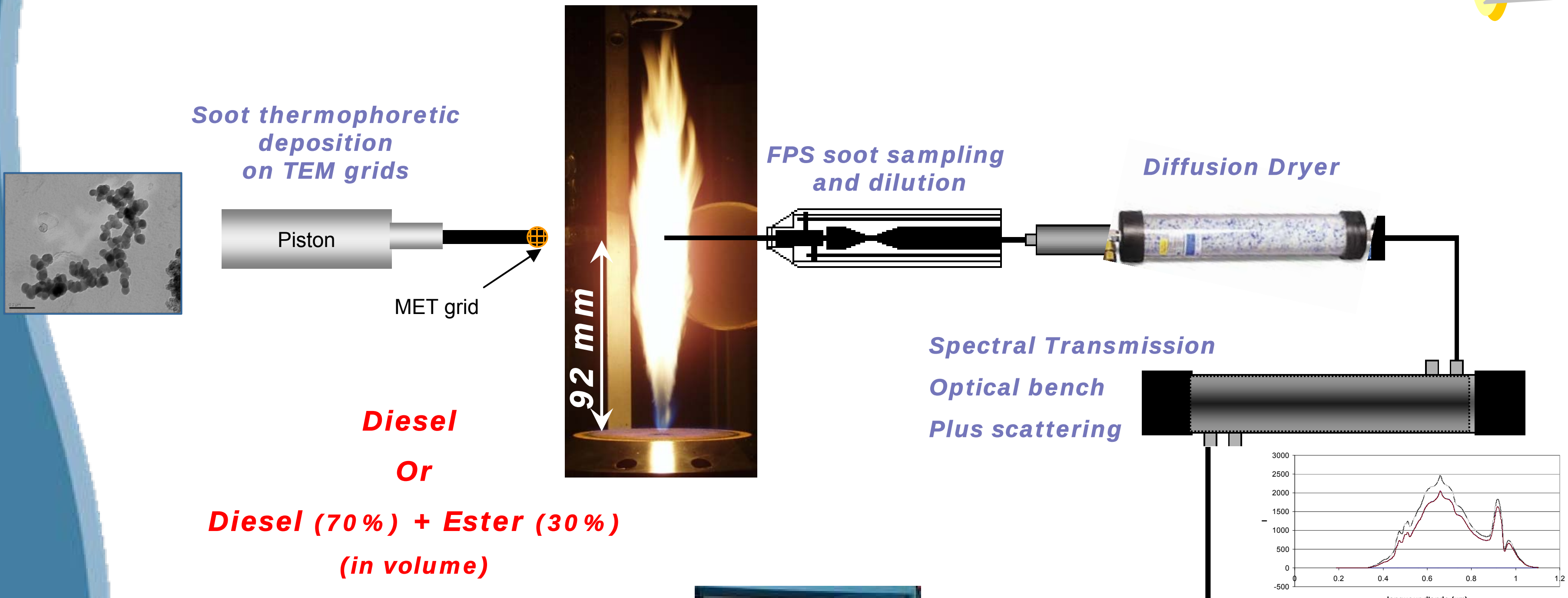
- Total uncertainty is on the order of 80-100%
- Diagnostic-specific uncertainty is on the order of 25-50%

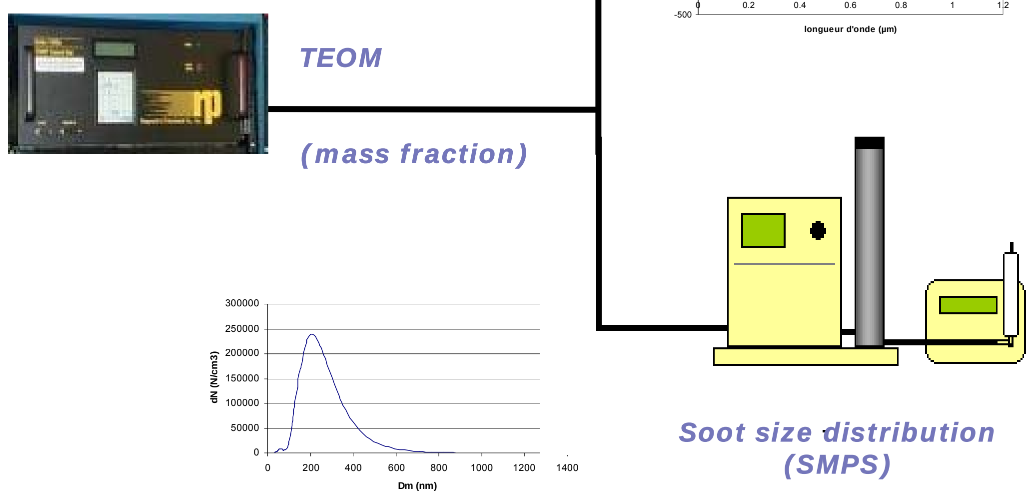
Determination of optical soot properties

- Using extinction and scattering measurements for soot sampled from flames operated with different liquid fuels
- Comparison to independent diagnostics

Experimental Setup

Second Approach : with light scattering



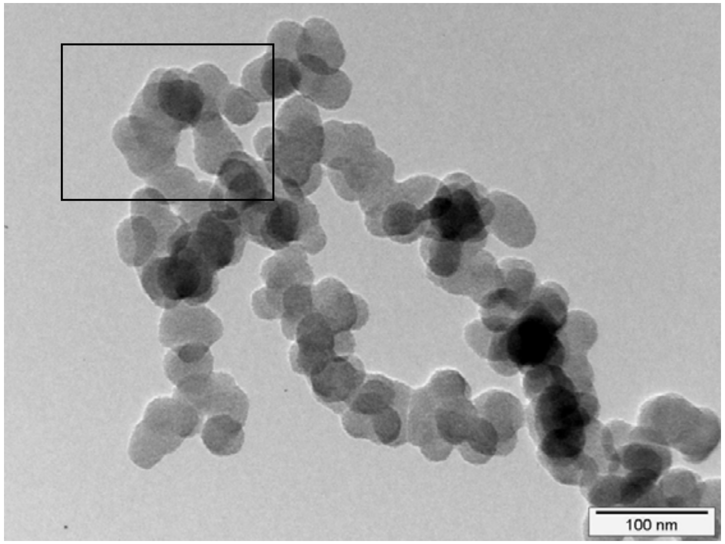


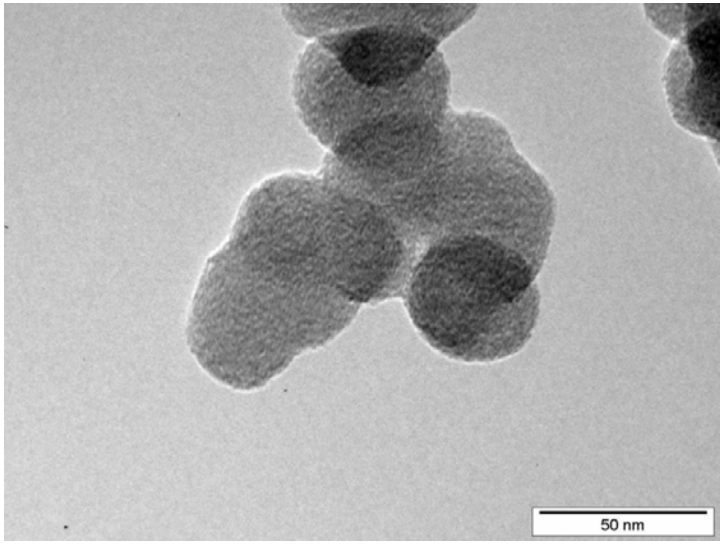
Setup allows for detailed study of morphology and its influence on the optical properties

Soot properties during the LII process

- The LII process is non-intrusive with respect to the total flame
- What is the influence on the soot particles?

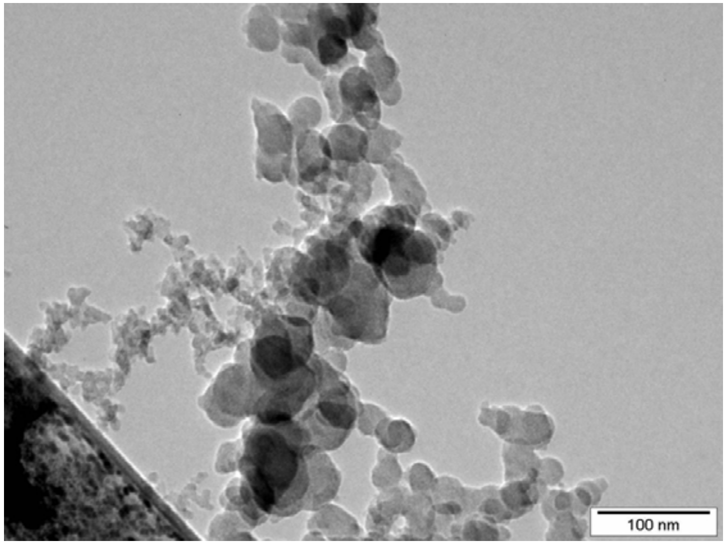
no laser

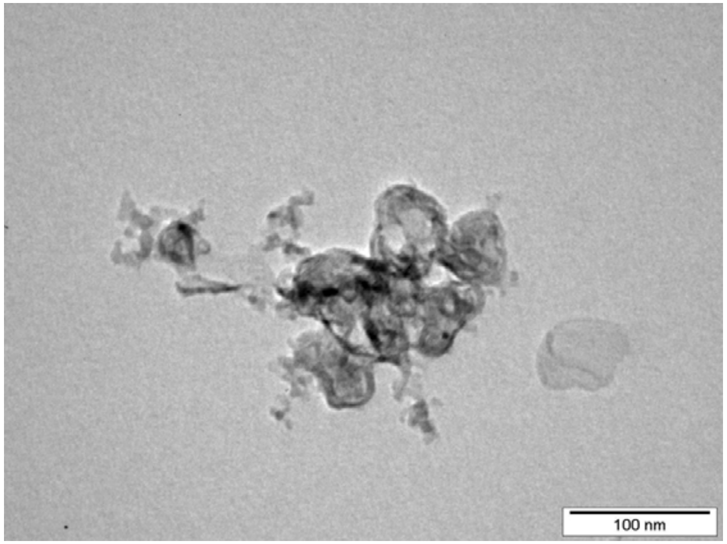


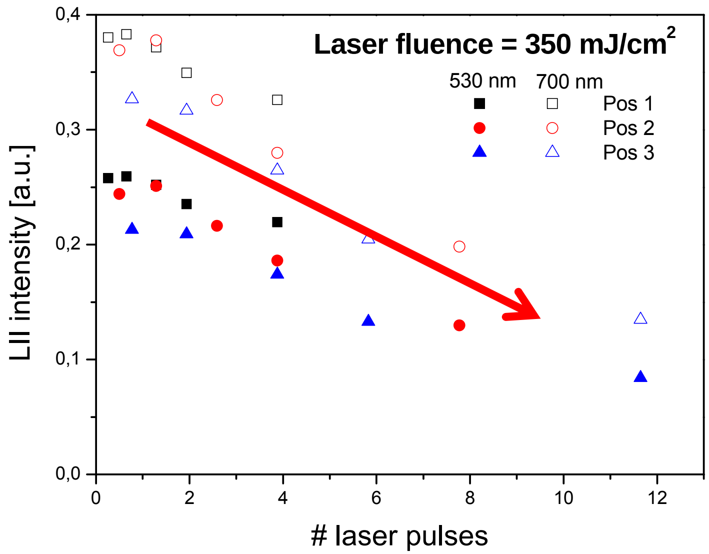


- TEM sampling in flame exhaust
- Monitoring LII signal emission in sampling pipe after different number of LII pulses

high fluence ~ 350 mJ/cm²

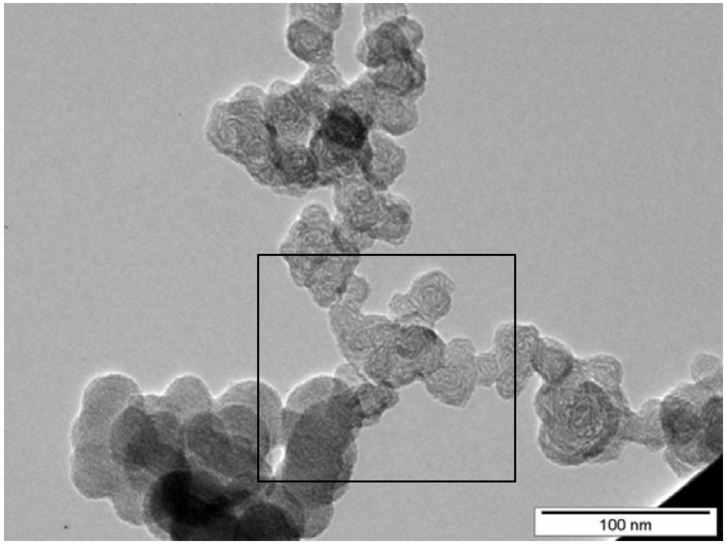


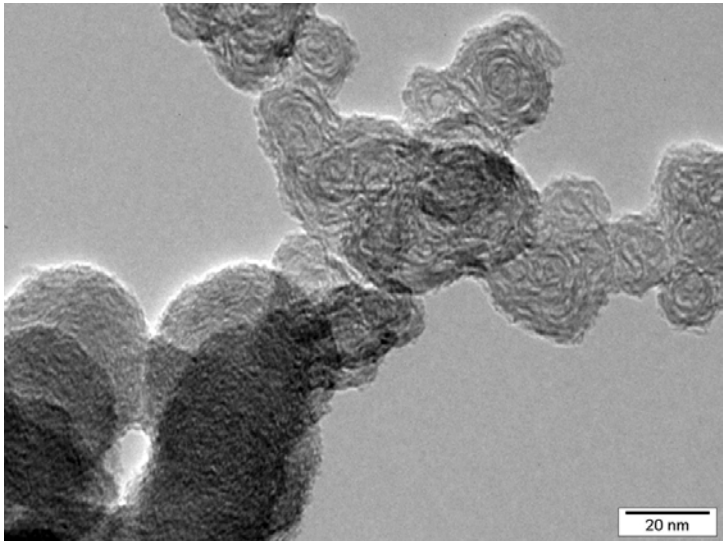


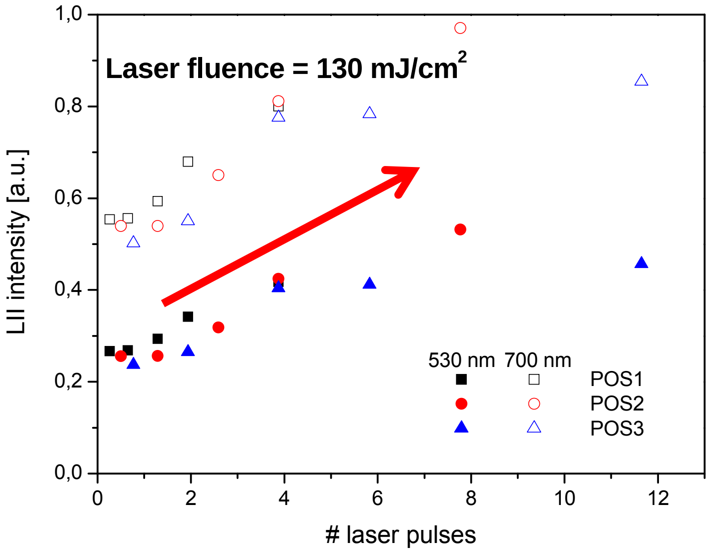


- High fluence is typically used for soot concentration mapping; LII signal is „proportional“ to f_v
- Soot is emitting LII signal before or during getting destroyed
- Accumulating laser pulses results in decreasing signal → surface vaporization

low fluence ~ 130 mJ/cm^2

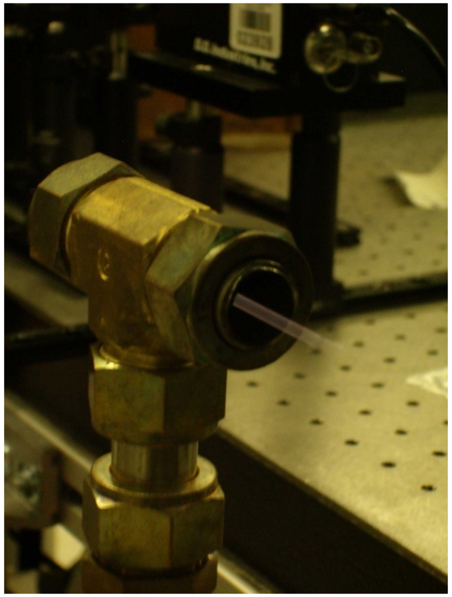






- Intermediate fluence is frequently used for particle sizing
- Moderate changes of soot morphology ($T_{peak} < T_{sub}$)
- Accumulating laser pulses results in increasing signal → T_{peak} increases with number of pulses; due to morphological changes allowing for better absorption?

Optical Properties of Soot – What happens during the LII laser pulse?



- Heat particles with IR pulse and study their absorptive behaviour continuously with any desired wavelength (cw)
- Pipe is flushed with soot from a quenched flame
- White (pulsed) LII emission is visible in picture

Session 3 - Combining LII with other diagnostics

- Scattering
- Extinction
- TEM

- To deduce soot morphological information

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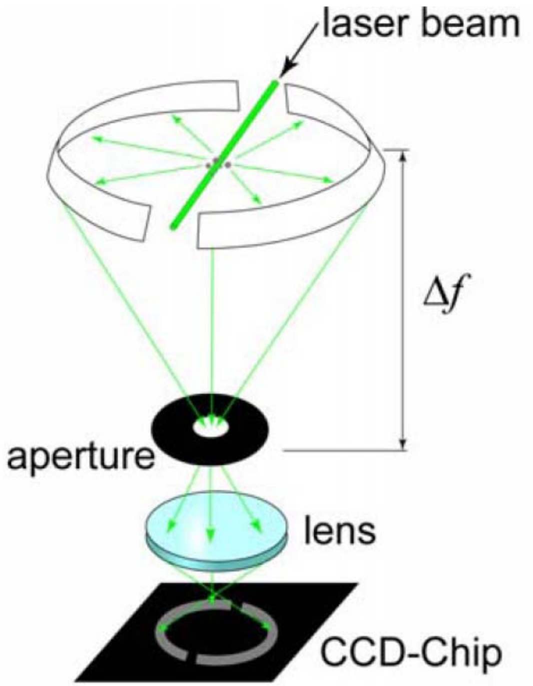
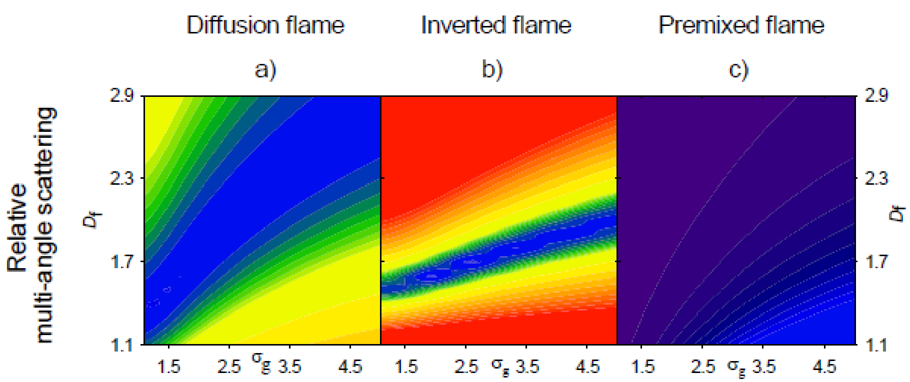
Wide or multi angle scattering and LII



- Angle range 10°-170°
- Applied to various flames
- Applied to SiO_2 and SnO_2 pyrolysis flames
- For the characterization of aggregates



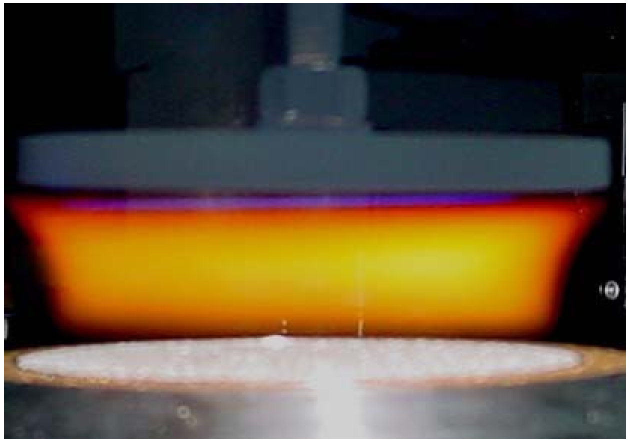
D_f:fractal dimension

σ_g:distribution width

Contributions by:
H. Oltmann et al., University Bremen
O. Link et al., NRC Ottawa

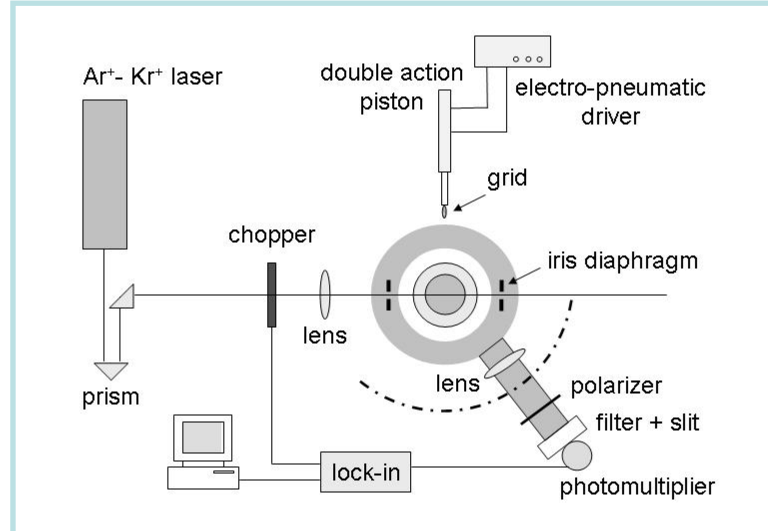
Combination of different diagnostics

- ✓ LII/extinction ⇨ f_v
- ✓ +Three-angle scattering ⇨ R_g, d_p
- ✓ TEM ⇨ D_f, K_f, R_g, d_p
- ✓ What about d_p derived from LII?

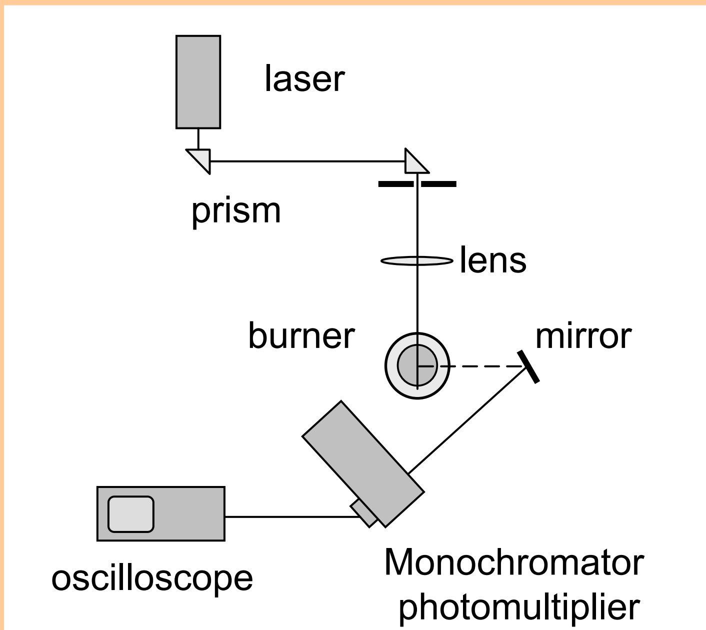


Premixed rich C_2H_4/air flame : Φ=2.34

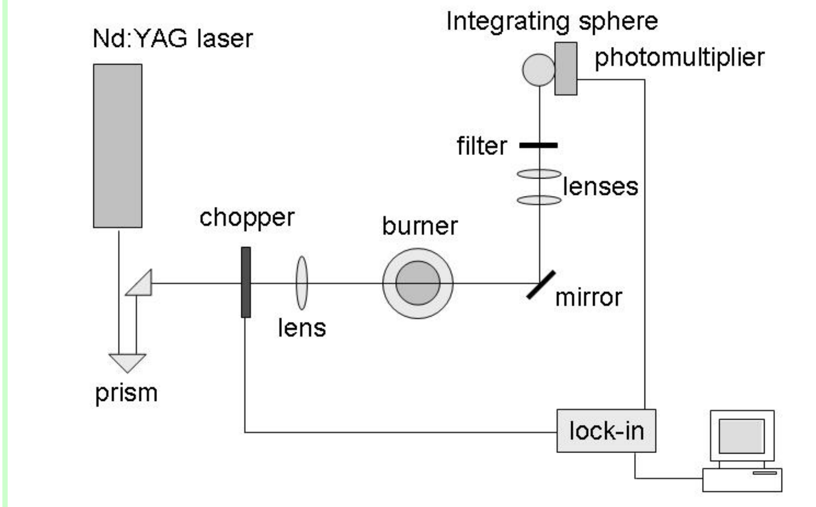
Multi-angle scattering/TEM



Two color LII set-up



Extinction



- Good agreement of TEM with optically determined parameters
  - Primary particle diameter
- Good agreement of 2C LII with extinction (at 1064 nm)
  - Soot concentration

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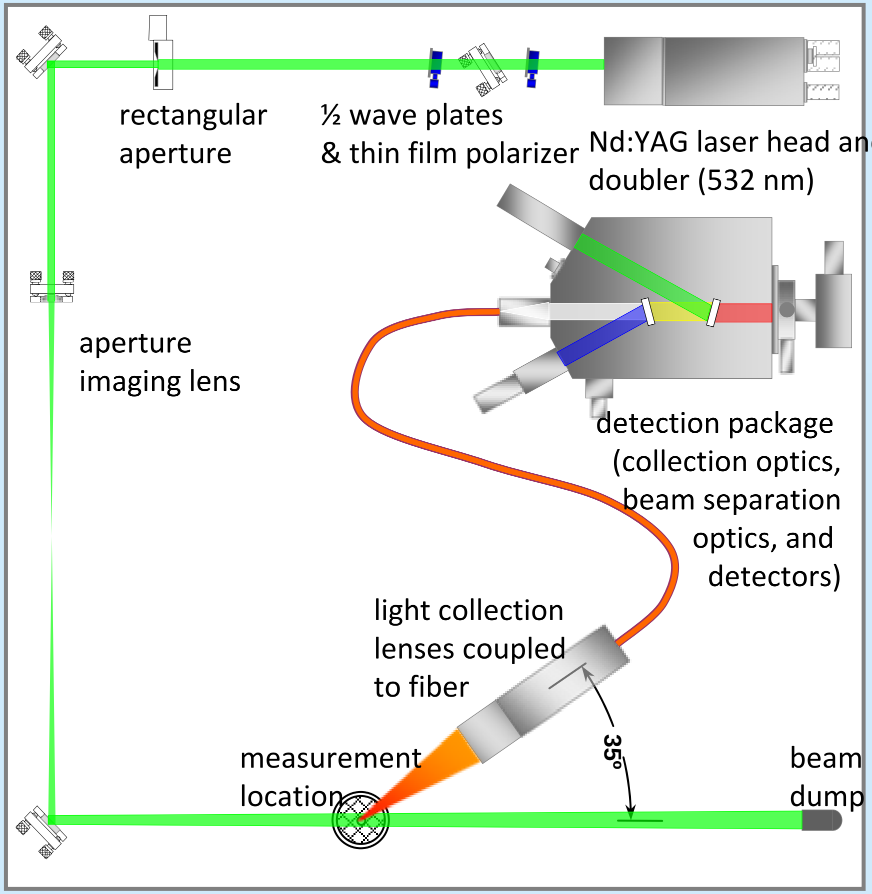
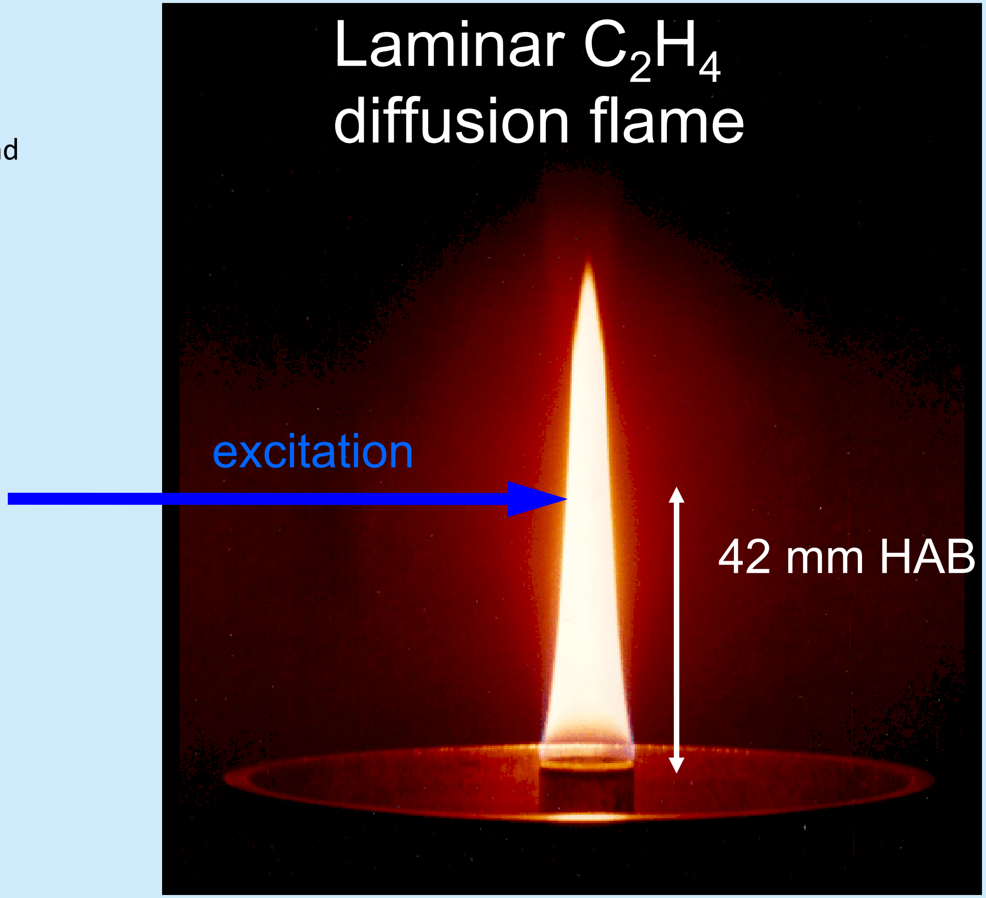
and Environmental
Technology

Simultaneous LII/scattering





Excitation 532 nm. LII & scattering data collected at 35 & 145 degrees in separate experiments

Detection : 450 nm Δλ=50 nm 780 nm Δλ=40 nm and 532 nm

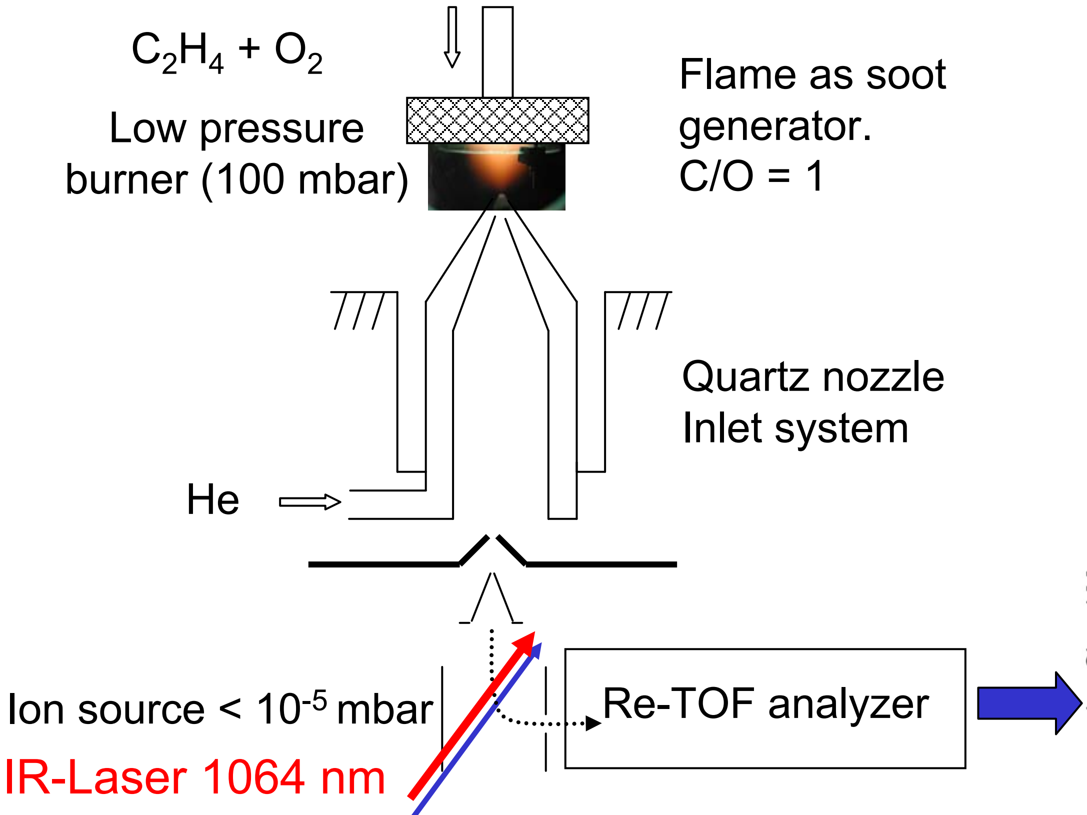
Combining LII with other diagnostics

- Time of flight mass spectrometry

- To determine species leaving the soot surface when intermediate to high fluence LII pulses are employed
  - So far, only assumptions or theoretical considerations are available
  - Neutral species of type C_3 to C_{10} are considered in LII models

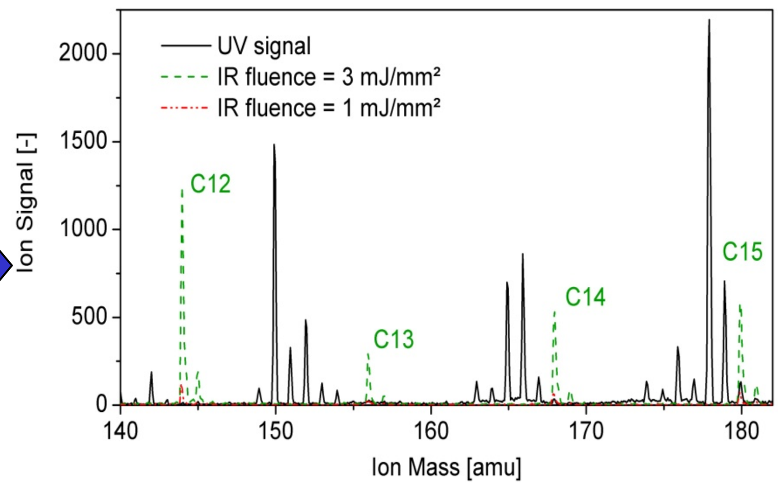
- Used pulsed UV laser fluences are known to be low enough not to fragment molecular and particular species
- Used pulsed IR laser is in the fluence range typical for LII (much higher fluence, but photons of much lower energy)

Experimental Set-Up - PIMS



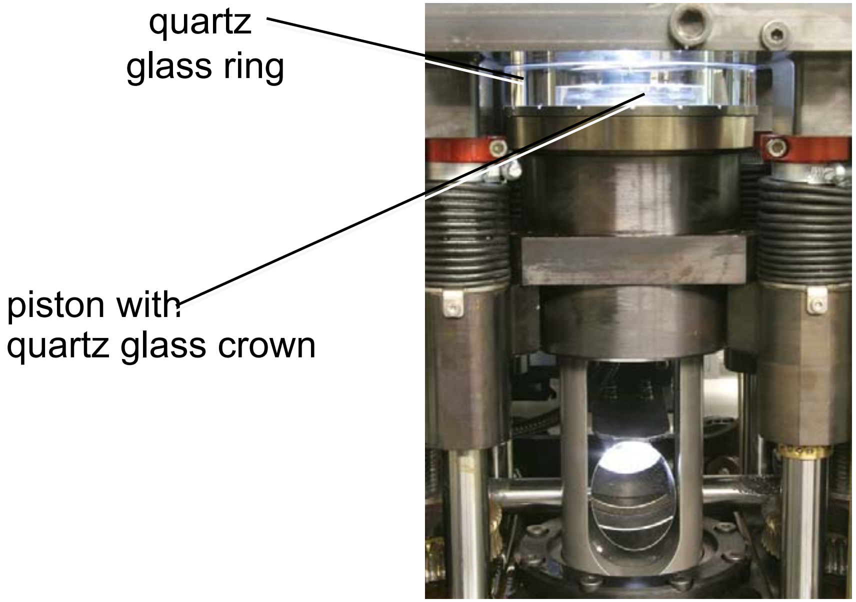
IR-Laser 1064 nm
and
UV Laser 193 nm

Small **ionic** C_n (n **up to 18**) clusters found with pulsed IR excitation, even without needing high energy UV photons

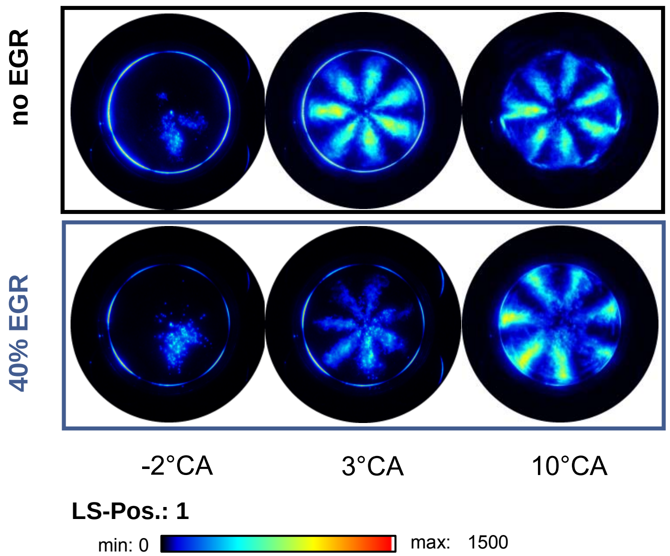


Session D - LII under various conditions

- Making use of an optically accessible engine
- Pure soot luminosity measurement (line of sight) was not sufficient to correlate with exhaust gas measurements when studying the influence of EGR
- LII allowed for qualitative and spatially resolved monitoring of soot fields over whole combustion cycle and for different EGR rates

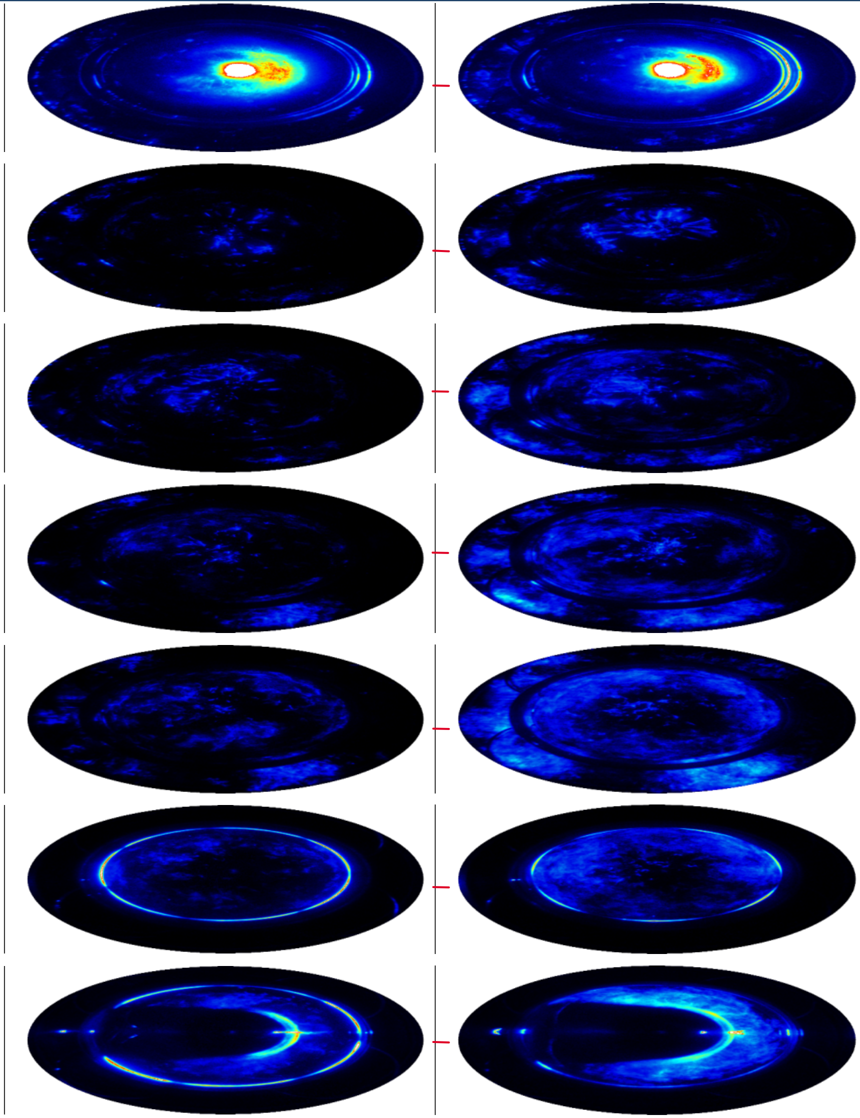


Examples of soot distributions during operation



BOSCH

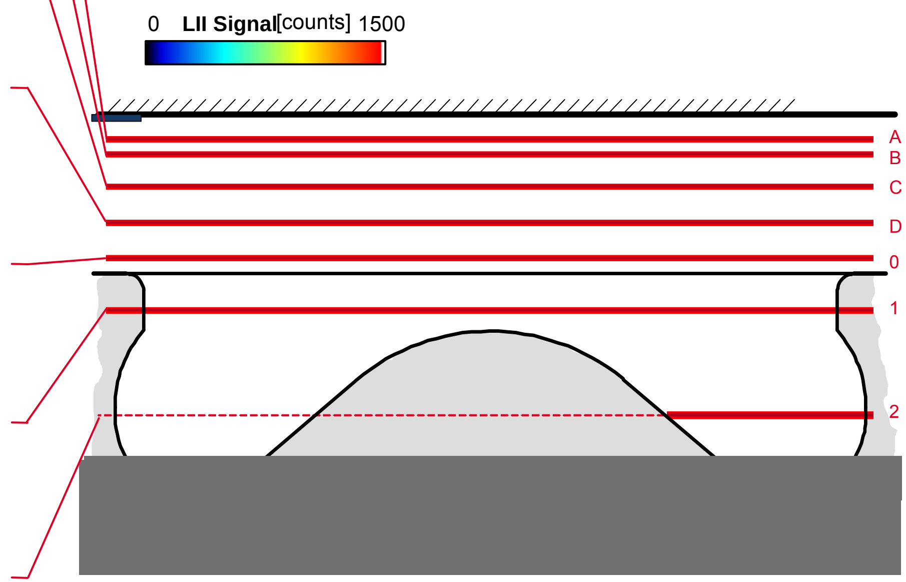
Results



Soot Distribution at 35°CA

left: no EGR, SZ = 0,1 FSN*
right: 40% EGR, SZ = 0,4 FSN*

* thermodynamic engine



LII in ultra high vacuum

- Negligible collisions of LII heated soot with ambient molecules
  → very long decay times on the order of 10s to 100s of µs (!)
  → lack of efficient particle cooling mechanisms
  → high detection sensitivity, allowing for studying single aggregates
  here: aged soot from particle generator

- New ultra-vacuum system has been designed
- Includes aerodynamic lens to bring particles into vacuum chamber
- CW Fiber laser for particle heating
- 2-colour pyrometric detection

- System assembled and currently being optimized

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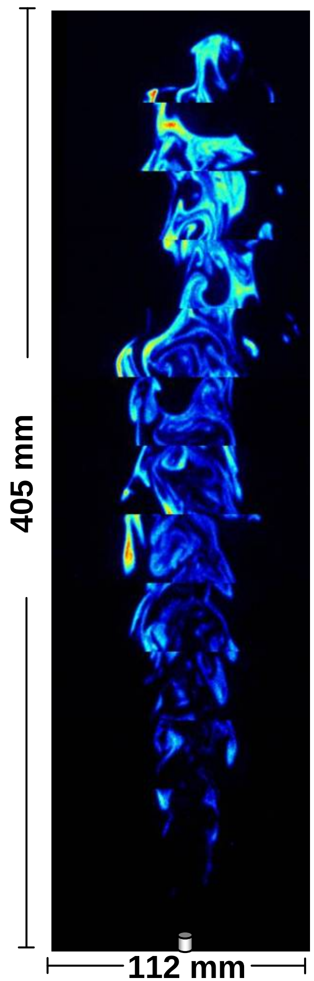
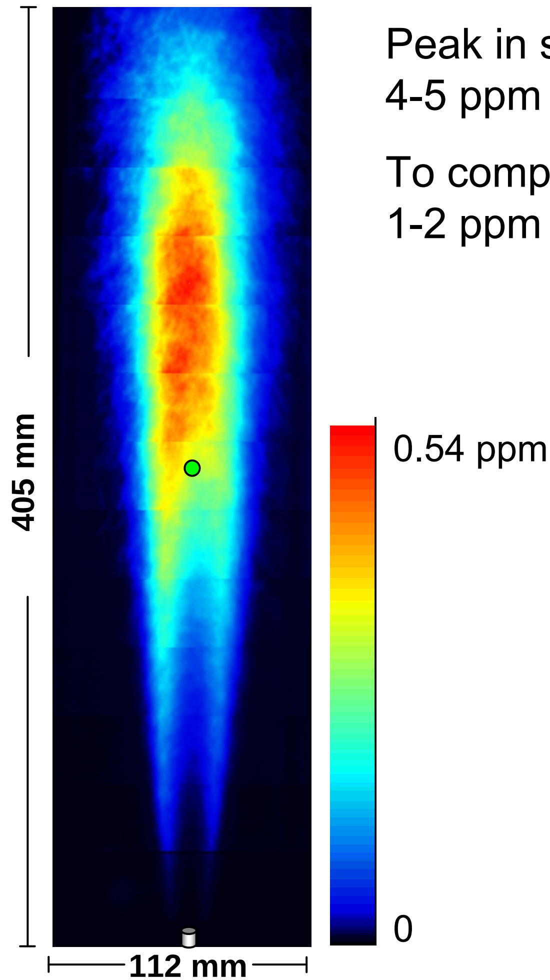
Adopt or optimize existing diagnostics

2D-LII



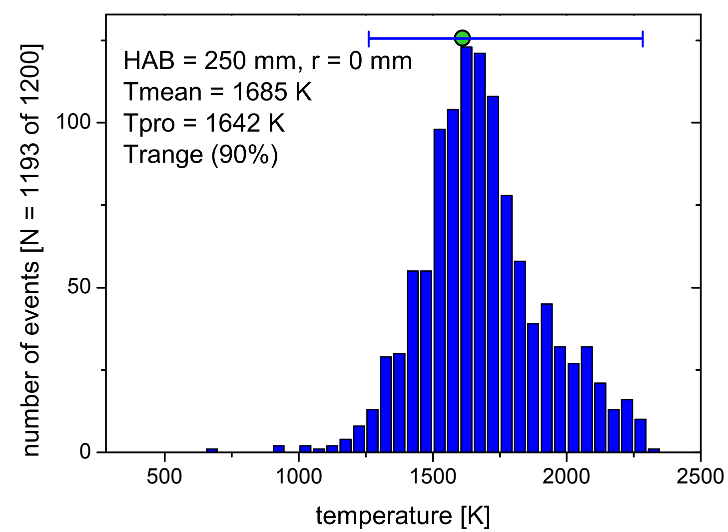
Soot concentration



Peak in single shot images:
4-5 ppm

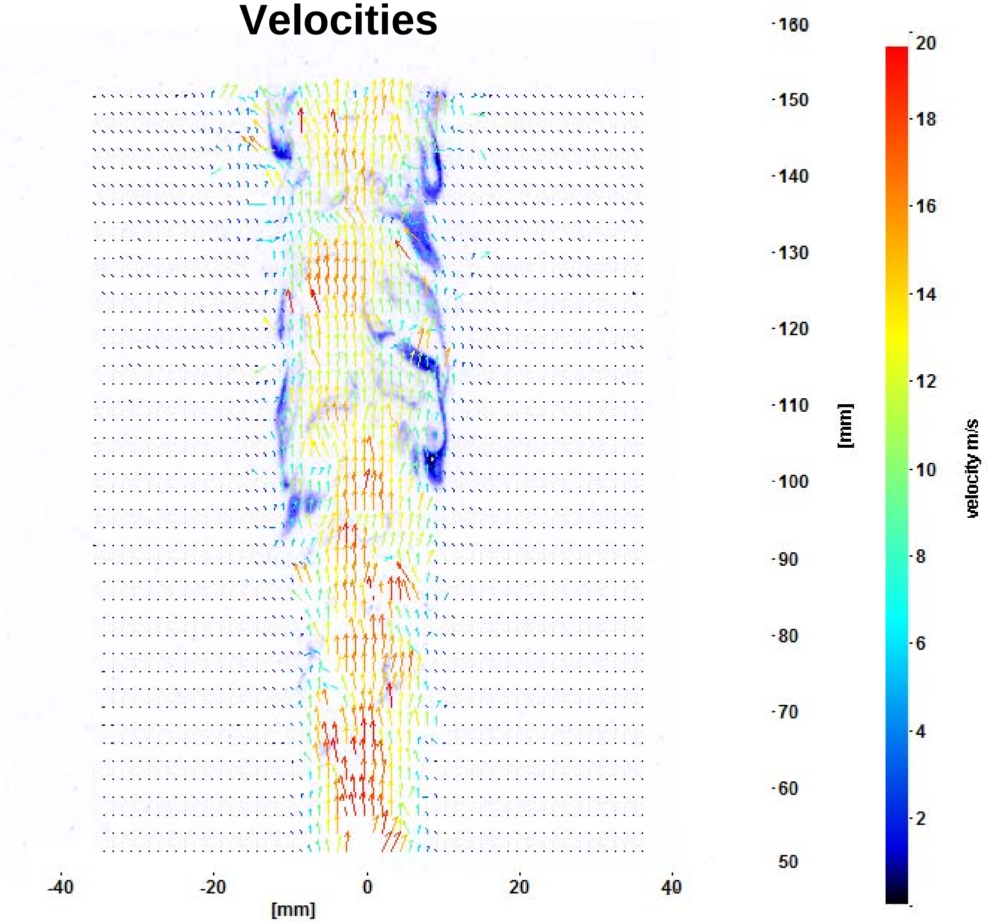
To compare with candle:
1-2 ppm

Temperatures



Adopt or optimize existing diagnostics

Velocities



- To date, PIV is not applied to sooting flames, despite the usefulness of this information → challenging task
- Data set is already being used by our CFD colleagues

Summary

- Framework of the presentation was insufficient to cover all topics in detail → rather some snapshots were shown indicating ongoing research
- Application of a variety of different complementary diagnostics supporting the understanding of LII and sooting flames
- Diagnostics development
- Synergies in the LII workshop community due to regular exchange of experts in the field
- Contributions to appear in near future Applied Physics B issue for those interested in more details
- Next Workshop Fall 2012, Lille, France

Recent IEA papers

2009

Scattering/extinction measurements for soot particle formation and growth in a shock tube *S. De Iuliis, G. Zizak, CNR-IENI, Italy, and N. Chaumeix, M. Idir, C.E. Paillard, CNRS-ICARE and Universitè d'Orléans, France*
Investigation of soot optical properties by spectral line-of-sight attenuation *Kevin Thomson, Francesca Migliorini, Greg Smallwood, National Research Council, Canada, and Klaus-Peter Geigle, DLR, Germany*
Investigation of diesel soot emissions during transient engine operation using in-cylinder pyrometry *Patrick Kirchen, Peter Obrecht, Konstantinos Boulouchos, ETH Zurich, Switzerland*
Complementary diagnostics for monitoring soot formation in fundamental flames by PIMS and LII and application of soot diagnostics to technical combustion *Klaus Peter Geigle, Jochen Zerbs, Horst-Henning Grotheer, Joachim Happold, DLR, Germany*
Progress on the design and construction of a high vacuum LII system using an aerodynamic lens *Douglas Greenhalgh, Vivien Beyer, Heriot-Watt University, UK*
Issues with applying LII to the measurement of nonvolatile particulate emissions *Greg Smallwood, Kevin Thomson, David Snelling, Fengshan Liu, National Research Council, Canada*
Characterization of silica and titania nanoparticles synthesized in a spray flame reactor *F. Cignoli, S. Maffi, C. Bellomunno, S. De Iuliis, G. Zizak, CNR-IENI, Italy*

2008

Small-Angle X-Ray and Laser Light Scattering from Fractal-Like Soot Clusters *S. di Stasio Italy*
Vacuum LII; New Directions for a Sensitive Nanoparticle Measurement Technology *D. Greenhalgh, UK*
Advanced Diagnostics for Nano-Sized Particles Produced in Flames *A. D'Alessio, A. D'Anna, P. Minutolo, and L. A. Sgro, Italy*
An Analytical Approach to the Detailed Analysis of Carbon Particulate Matter *A. Ciajolo, R. Barbella, A. Tregrossi, B. Apicella, and M. Alfè, Italy*
Nanoparticles Characterization in the Cylinder and in the Exhaust of Internal Combustion Engines *B. M. Vaglieco, Italy*
Characterization of Titania Nanoparticles Synthesized in a Flame *C. Bellomunno, F. Cignoli, S. De Iuliis, S. Maffi, and G. Zizak, Italy*
In-situ Characterization of Particle Formation Processes with Subnanometer Structural Resolution on Nanoparticles and Soot (Size, Structure and Dynamics) Using X-ray Scattering Techniques *F. Ossler, S. Canton, J. Larsson, Sweden*
Detection of Fluorescent Nanoparticles in Flames with Femtosecond Laser-Induced Fluorescence Anisotropy *A. Bruno, F. Ossler, C. de Lisio, P. Minutolo, N. Spinelli, A. D'Alessio, Italy/Sweden*

In red: groups regularly contributing to LII workshops

Laser-induced incandescence for particle size and soot volume fraction

$$-4\pi r^2 \Lambda (T - T_0)$$

Heat transfer

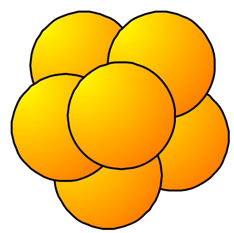
$$-\frac{\Delta H_V}{M} \cdot \frac{dm}{dt}$$

Vaporisation

~ Soot concentration

$$+\pi r^2 K_{abs} \cdot W_l$$

Absorption



Radiation

$$-\bar{\varepsilon} \cdot \sigma_{SB} (T^4 - T_0^4) \cdot 4\pi r^2$$

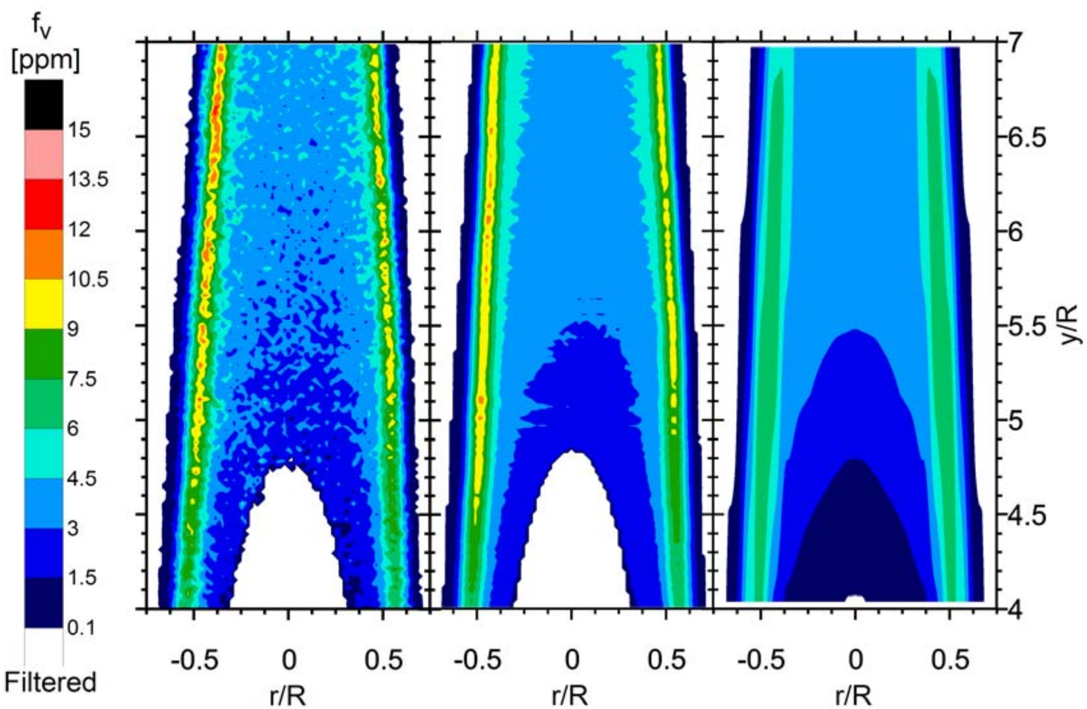
$$-\tfrac{4}{3}\pi r^3 \sigma \cdot C \cdot \frac{dT}{dt}$$

Internal energy

- Oxidation
- Annealing
- Others

Soot Volume Fraction Results

- Instantaneous measurements via 2D-AC-LII
- Averaged 2D-AC-LII measurements
- Comparing to 2D-LOSA measurements (Trottier et al)



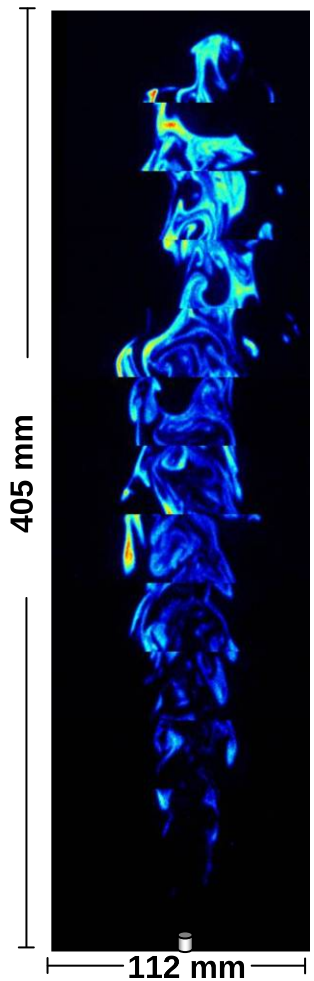
- These experiments served as starting point for determining experimental uncertainty
- Not detailed here

Supporting soot modellers

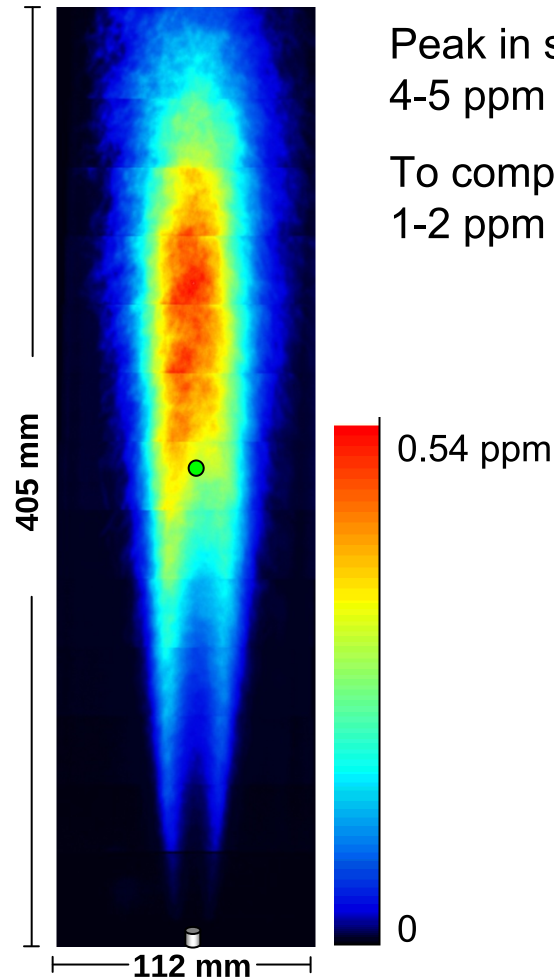
- Simple, well defined flame
- Technically relevant features (high turbulence)
- Bundle of precise non-intrusive diagnostics
- …

Adopt or optimize existing diagnostics

2D-LII



Soot concentration



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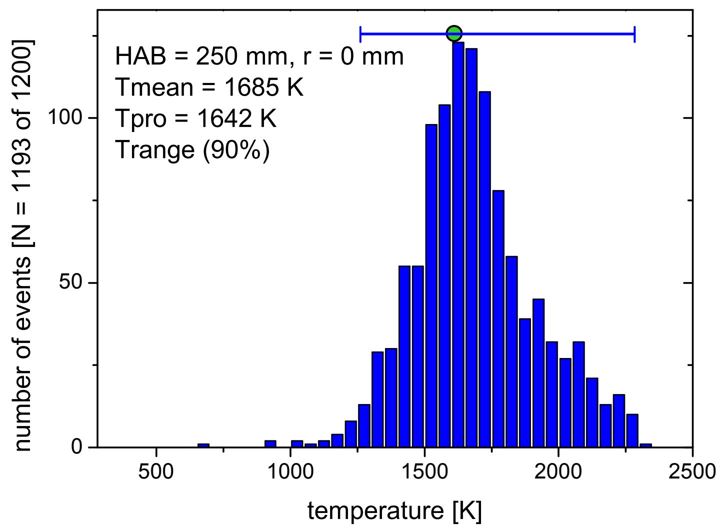
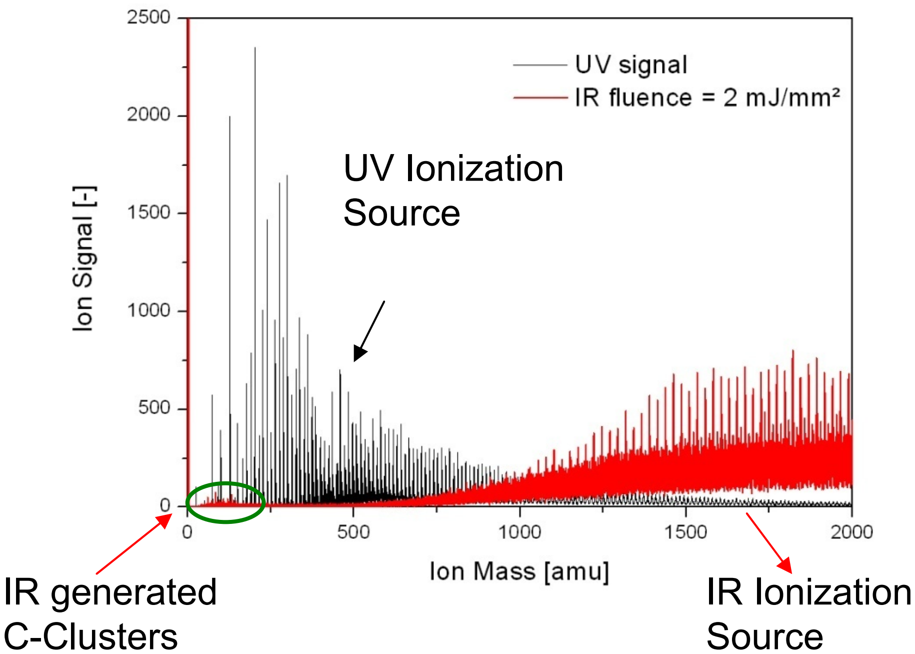
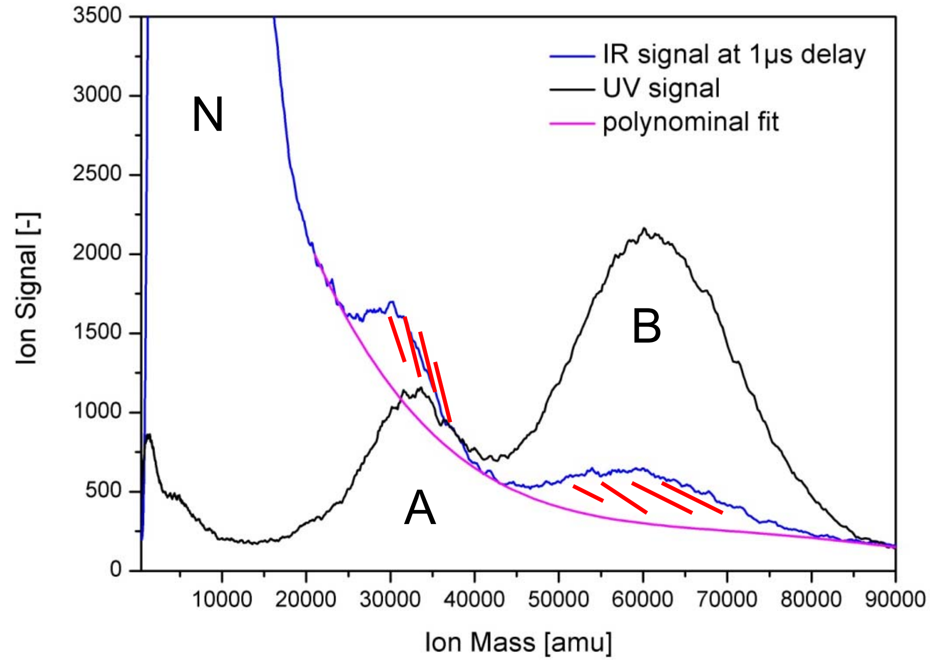
Temperatures



Results- Particle Phase (500 - 100.000 amu): Delay





- **Striking new feature: new particle mode (N) around 3000 u (i.e. approx. 1.5 nm)**
- A and B mostly destroyed by IR (2 mJ/mm²)
  B more than A
- IR residues are mostly charged
- Additional UV yields no signal enhancement
  not shown in plot

Interpretation so far:
complete graphite layers

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