4. ”Semi-intrusive” techniques

I. Measurements of soot - Laser induced incandescence, LII

II. Laser-induced breakdown spectroscopy, LIBS

I. Optical diagnostics of soot in flames

- **Soot formation**

- **Soot measurements using scattering/extinction**

- **Soot measurements using Laser-Induced Incandescence, LII**

Soot in combustion processes

- For health and environment hazardous emissions
- Increased radiation, more effective heat transfer.
  Important in boilers, furnaces, camp fires,
  candles etc. crucial component in fire spread
- Incomplete combustion, reduced engine efficiency
- Increased wear leading to reduced lifetimes of components
- Deposits in engines, turbines, furnaces

Engine Emissions

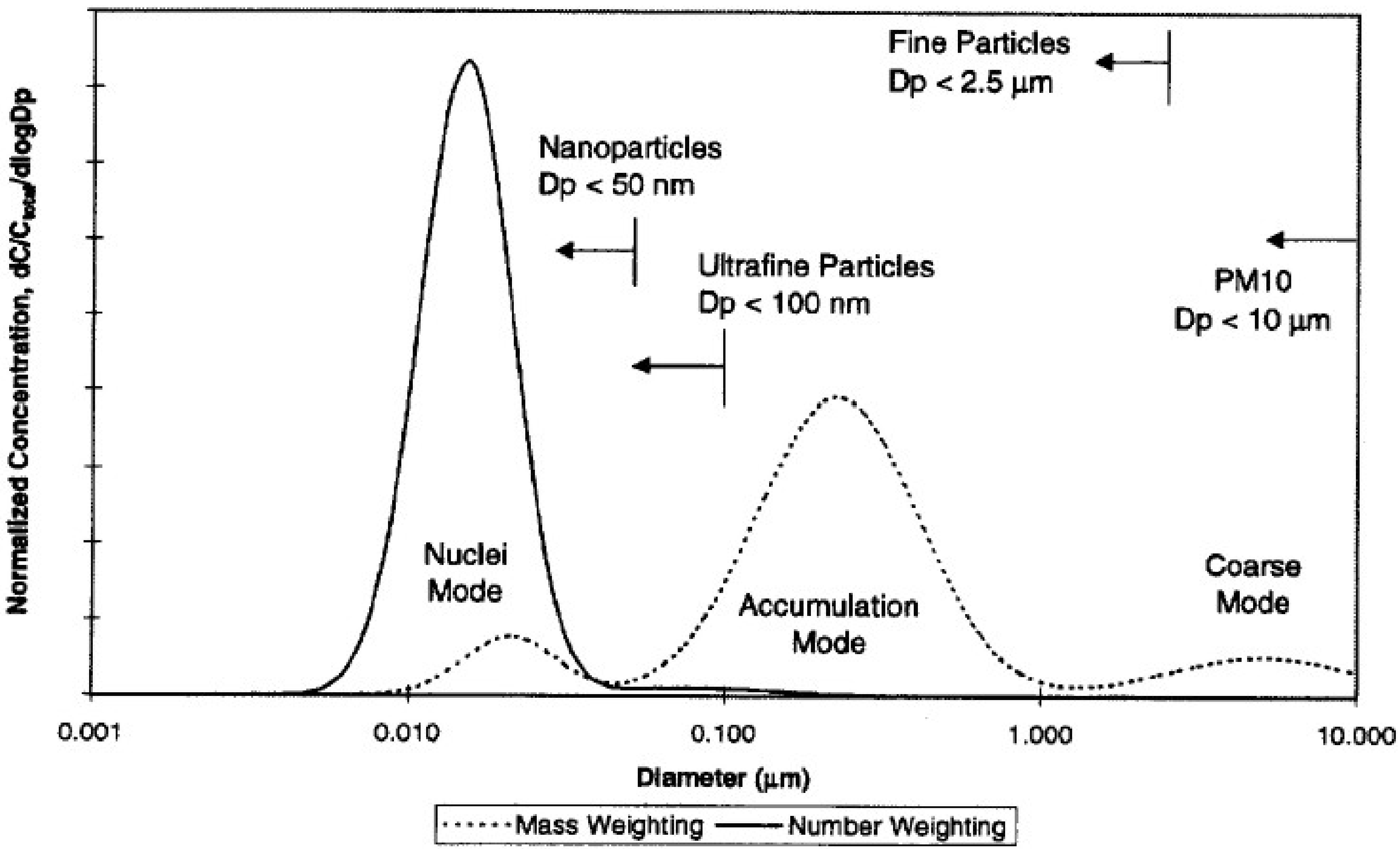
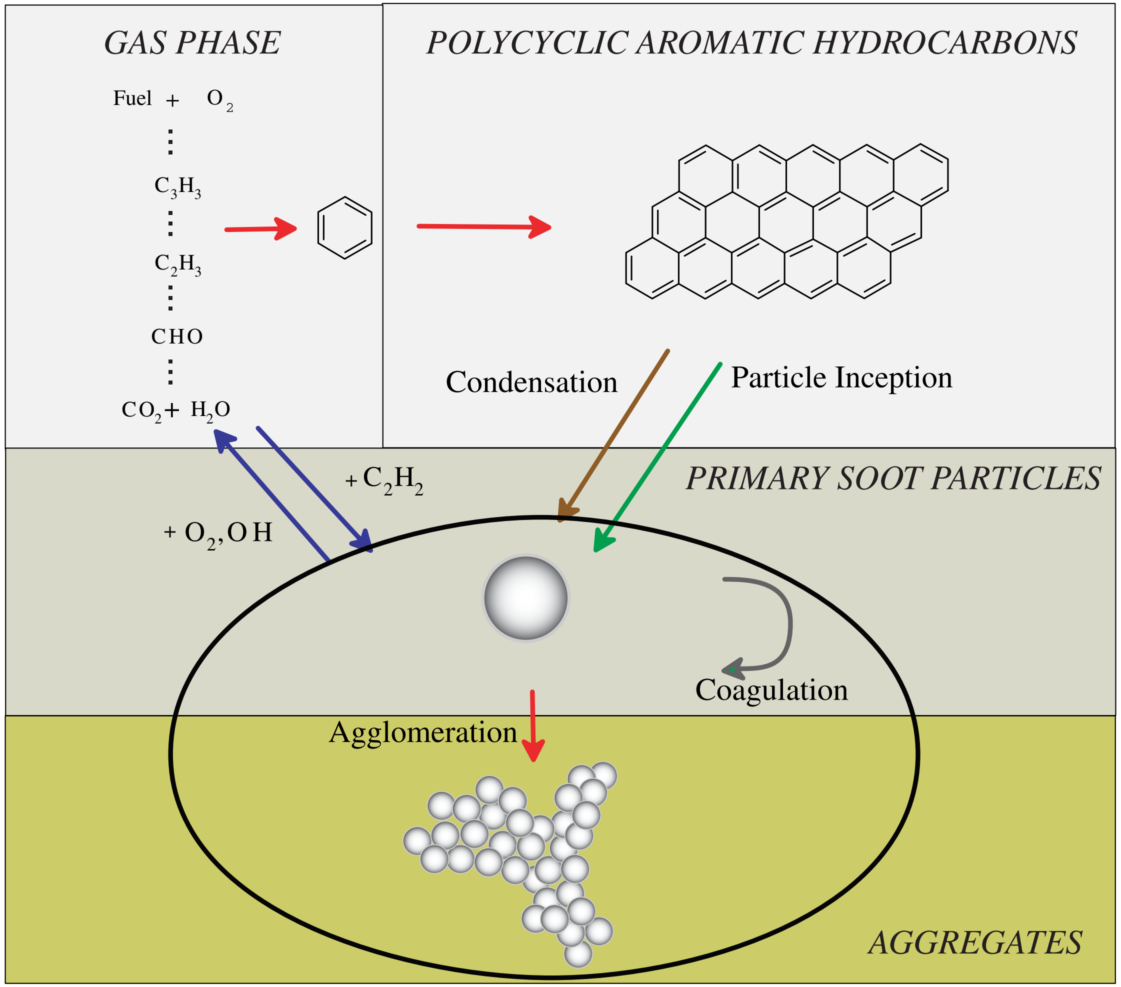


Fig. 3. Typical engine exhaust size distribution both mass and number weightings are shown.

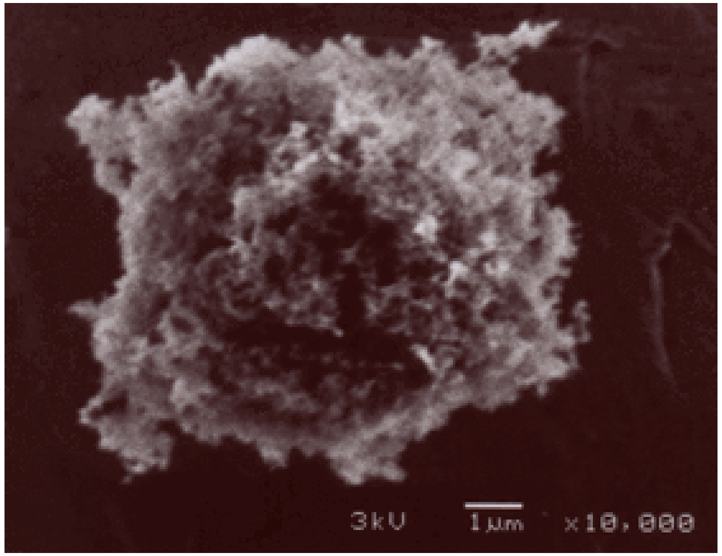
Kittleson (1998)

Soot formation



Soot characteristics

Primary soot particles
10-40nm



Black body radiation

Black body radiation has a continuous spectrum described by Planck's radiation law

$$I(\lambda) = \frac{2\pi hc^2}{\lambda^5} \frac{1}{e^{hc/\lambda kT} - 1}$$

$\varepsilon(\lambda)$ – emissivity !

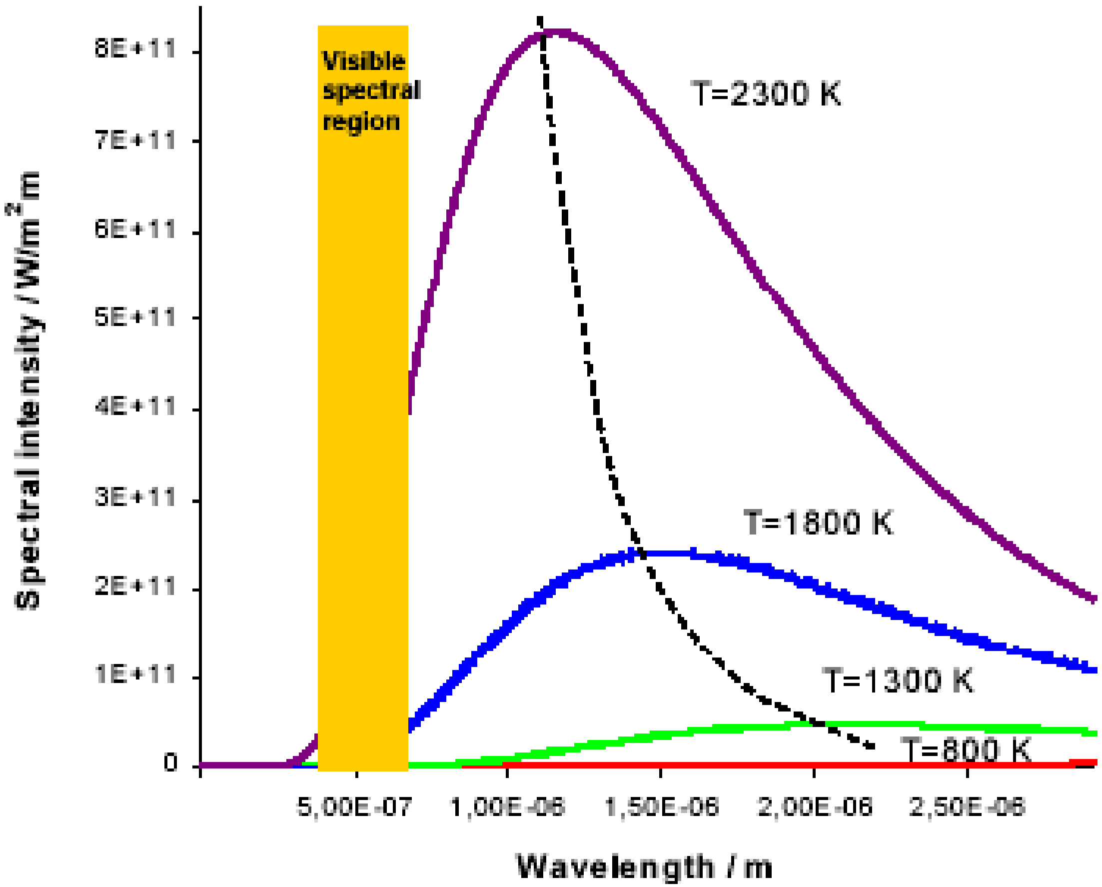
The radiation has a maximum that is shifted to shorter wavelengths with increased temperature. This is described by Wien's displacement law:

$$\lambda_{max} T = 2{,}898 \cdot 10^{-3}\,\mathrm{K} \cdot \mathrm{m}$$

The total intensity of the radiation is increasing with temperature according to Stefan-Boltzmann's law:

$$I = \sigma T^4$$

Planck emission from soot particles



Soot extinction

Assuming d << λ

$$I_0/I_T = \exp(LK_{ext})$$

$$K_{ext} = -\frac{\pi^2}{\lambda} \cdot \mathrm{Im}\left\{\frac{m^2-1}{m^2+2}\right\} \cdot N \cdot d^3$$

N; number of soot particles

d; particle diameter

m; complex refractive index of soot particle

Conclusion:

Soot volume fraction measurable

Line of sight technique

No size information

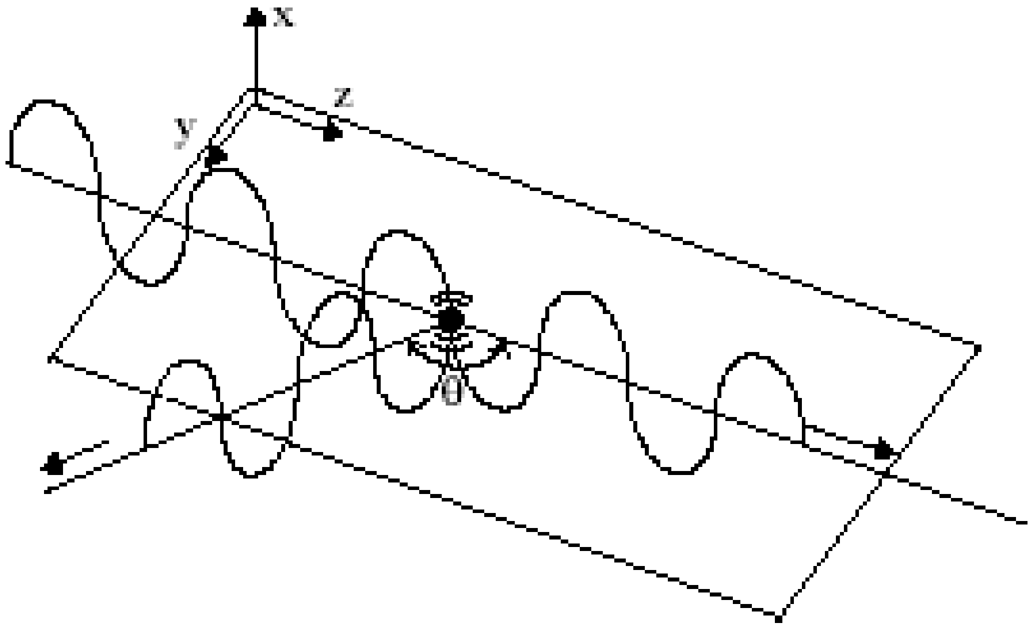
Soot scattering

Pol. of laser light

Soot scattering;

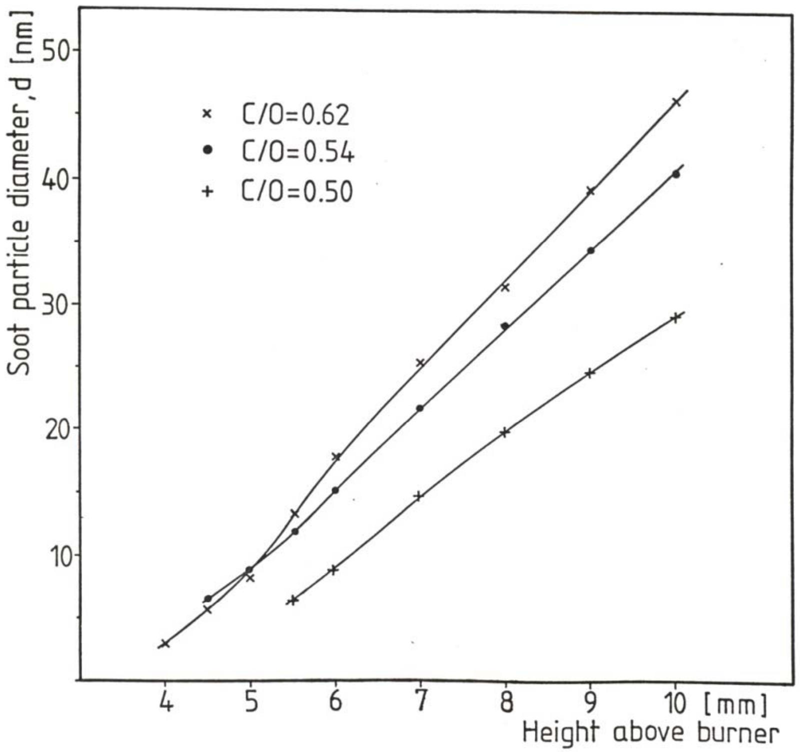
$$Q_{vv} = \frac{\pi^4}{4\lambda^4}\left|\frac{m^2-1}{m^2+2}\right|^2 Nd^6$$

Pol. of the scattered light



Soot scattering/extinction

Combined scattering /extinction can be used for measurements of N and d

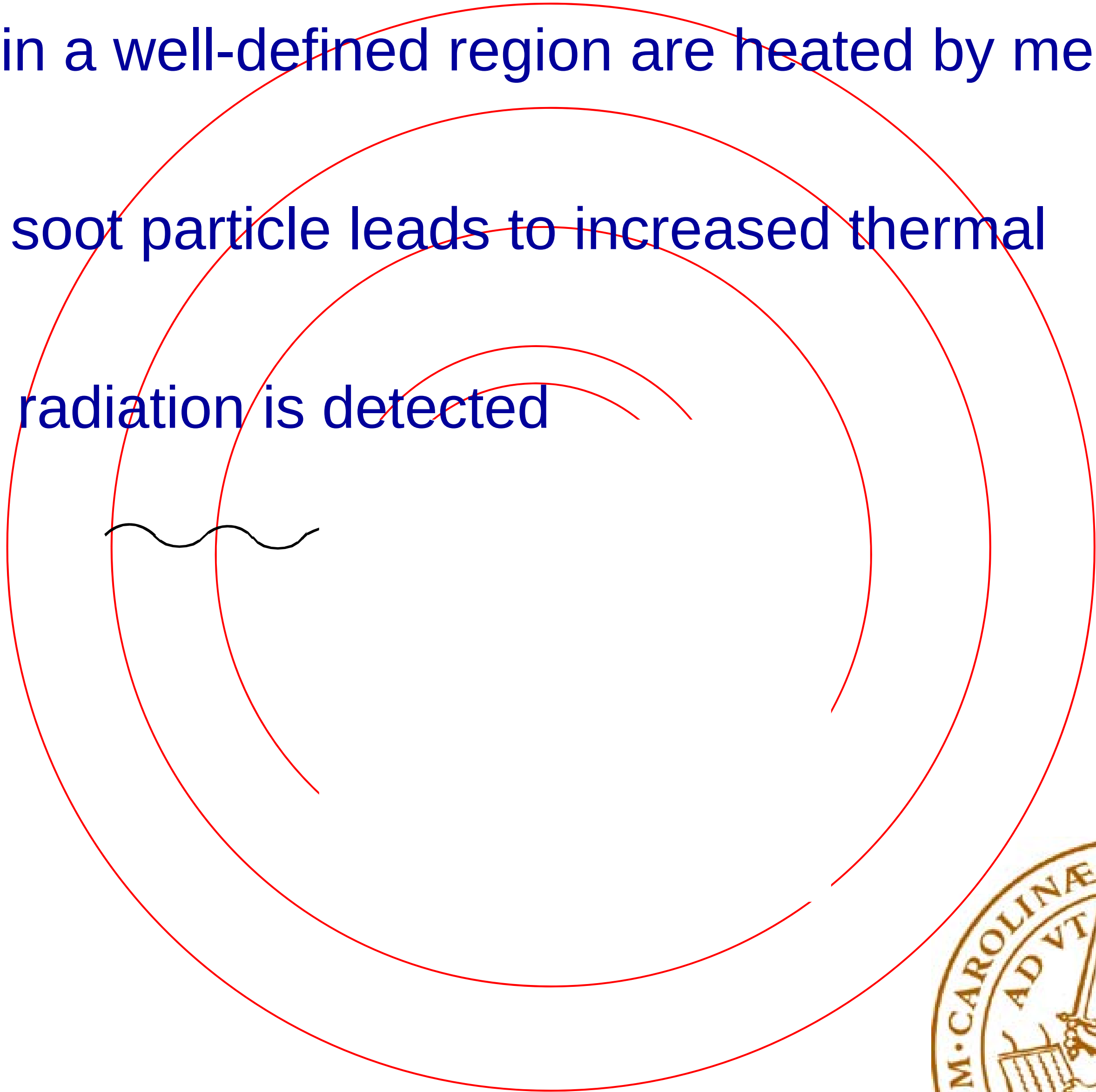


Laser-Induced Incandescence, LII

Soot particles in a well-defined region are heated by means of laser radiation

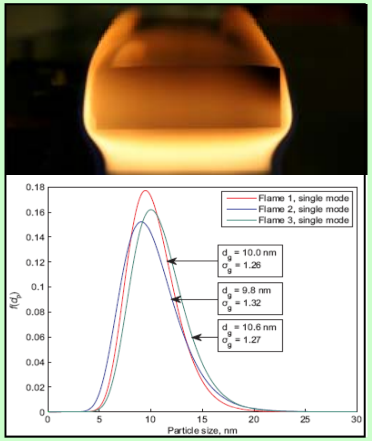
Heating of the soot particle leads to increased thermal radiation

The increased radiation is detected



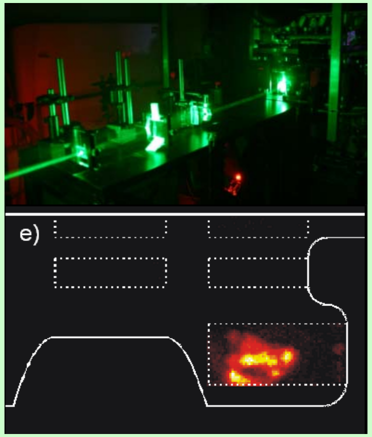
Applications of laser-induced incandescence

Flame studies



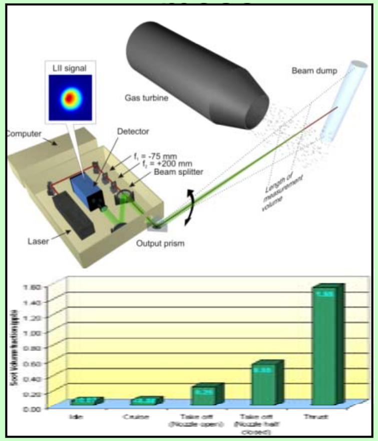
Johnsson et al. 2008 (poster)

In-cylinder engine



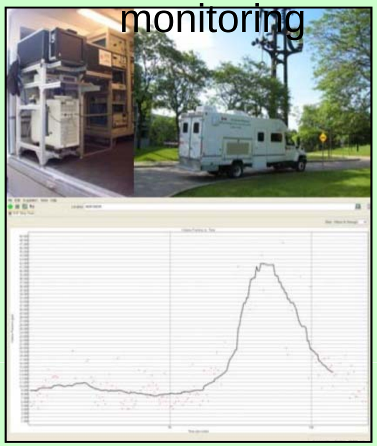
Bladh et al. 2006

Aero-engine exhaust



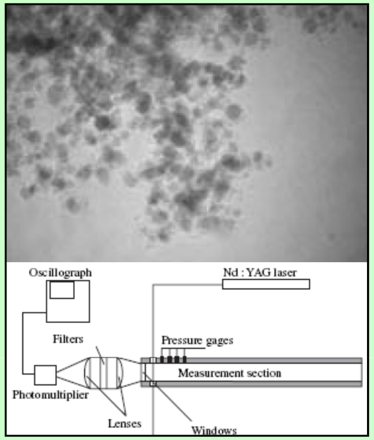
Desgroux et al. 2008

Environmental monitoring



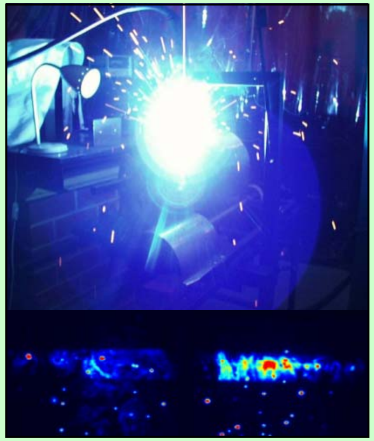
Smallwood et al. 2006

Nanoparticle charact.



Gurentsov et al. 2005

Welding fumes



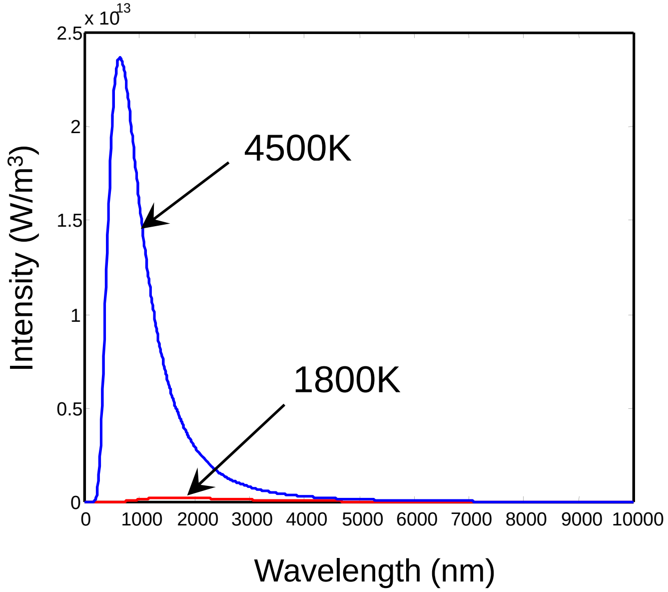
Lucas et al. 2006

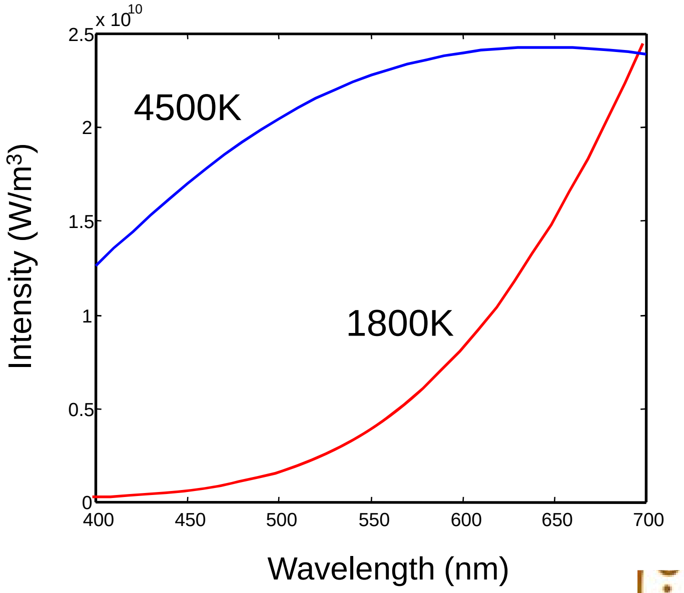
Courtesy: Henrik Bladh

Increased black body radiation by LII

The red curve shows the black body radiation as a function of the wavelength for T=1800 K, which is a typical flame temperature. The blue curve shows the same radiation for T=4500 K, which is within the same temperature range as a laser heated soot particle.

The left figure shows the real intensity difference for a large wavelength interval, while the right figure shows the normalised signal strength for visible light (normalization factor ~950).



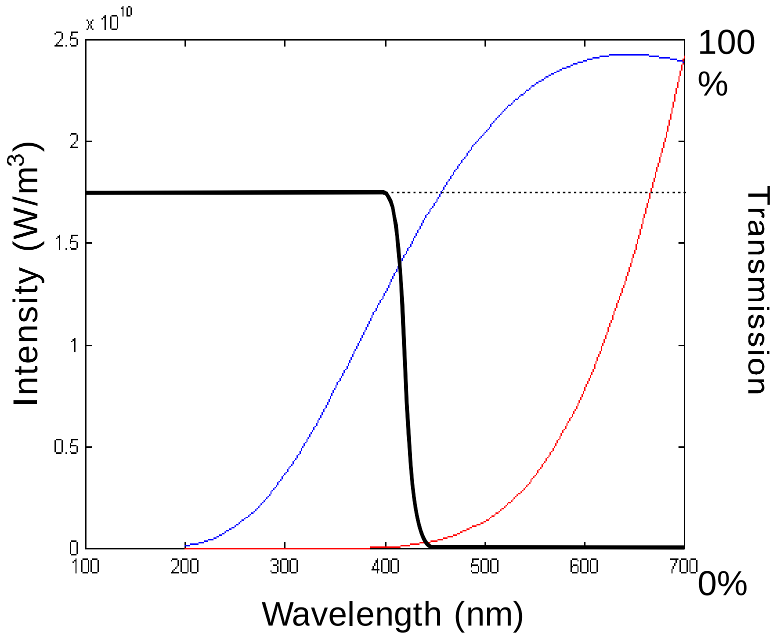


Background and LII

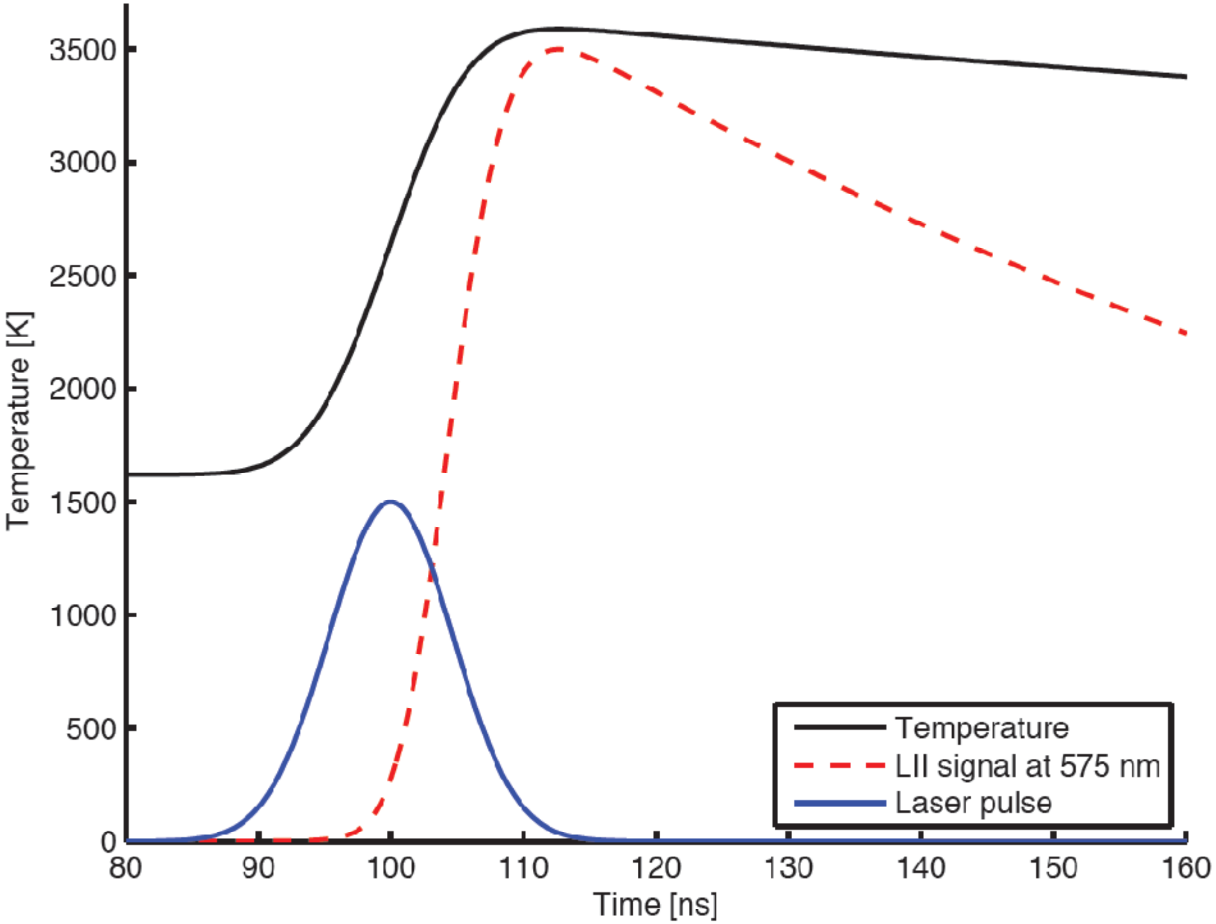
Spectral behavior:

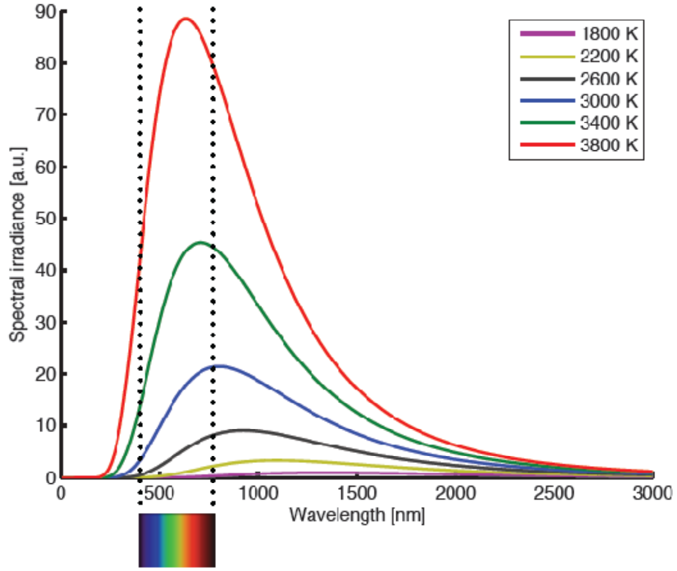
A spectral short-pass filter can be used to suppress longer wavelengths. The laser-heated soot radiates more at shorter wavelengths than the flame does.

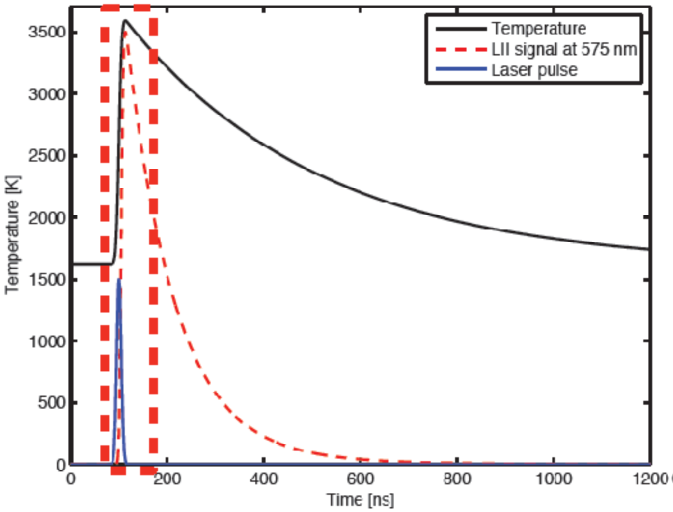
Wien's displacement law!



Soot temperature and LII signal



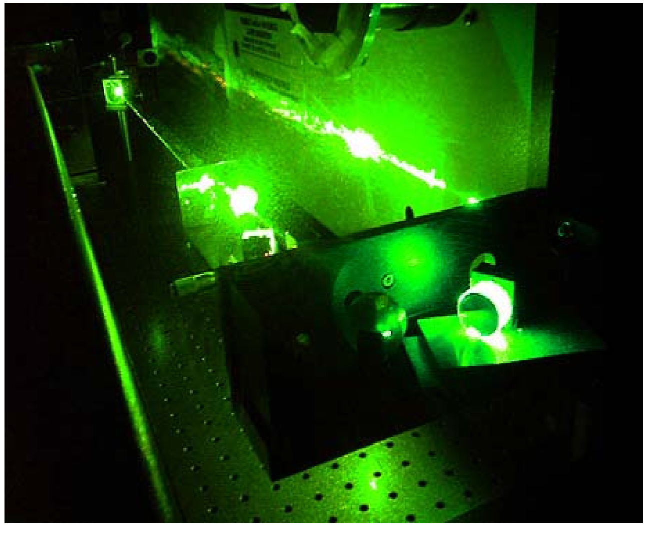




LII - The laser wavelength

Which wavelength is suitable for LII?

Soot absorbs well in a large wavelength interval (UV - visible – IR). There are, however, other species that can be excited with UV light and cause disturbing fluorescence. Therefore wavelengths in the UV should not be used. Most common is the Nd:YAG laser at 1064 nm (IR) and its second harmonic at 532 nm.

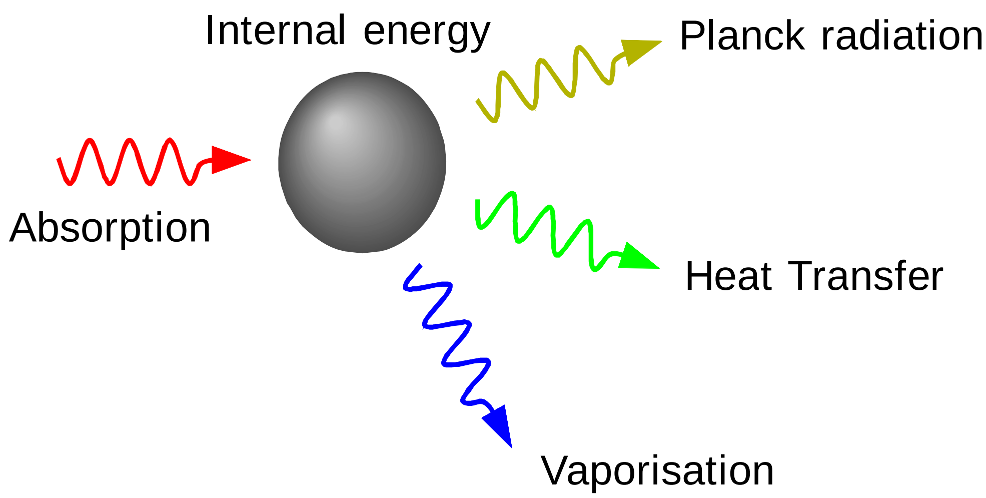


The radiation-particle interaction

A soot particle is exposed to laser radiation.

The following processes are intitiated

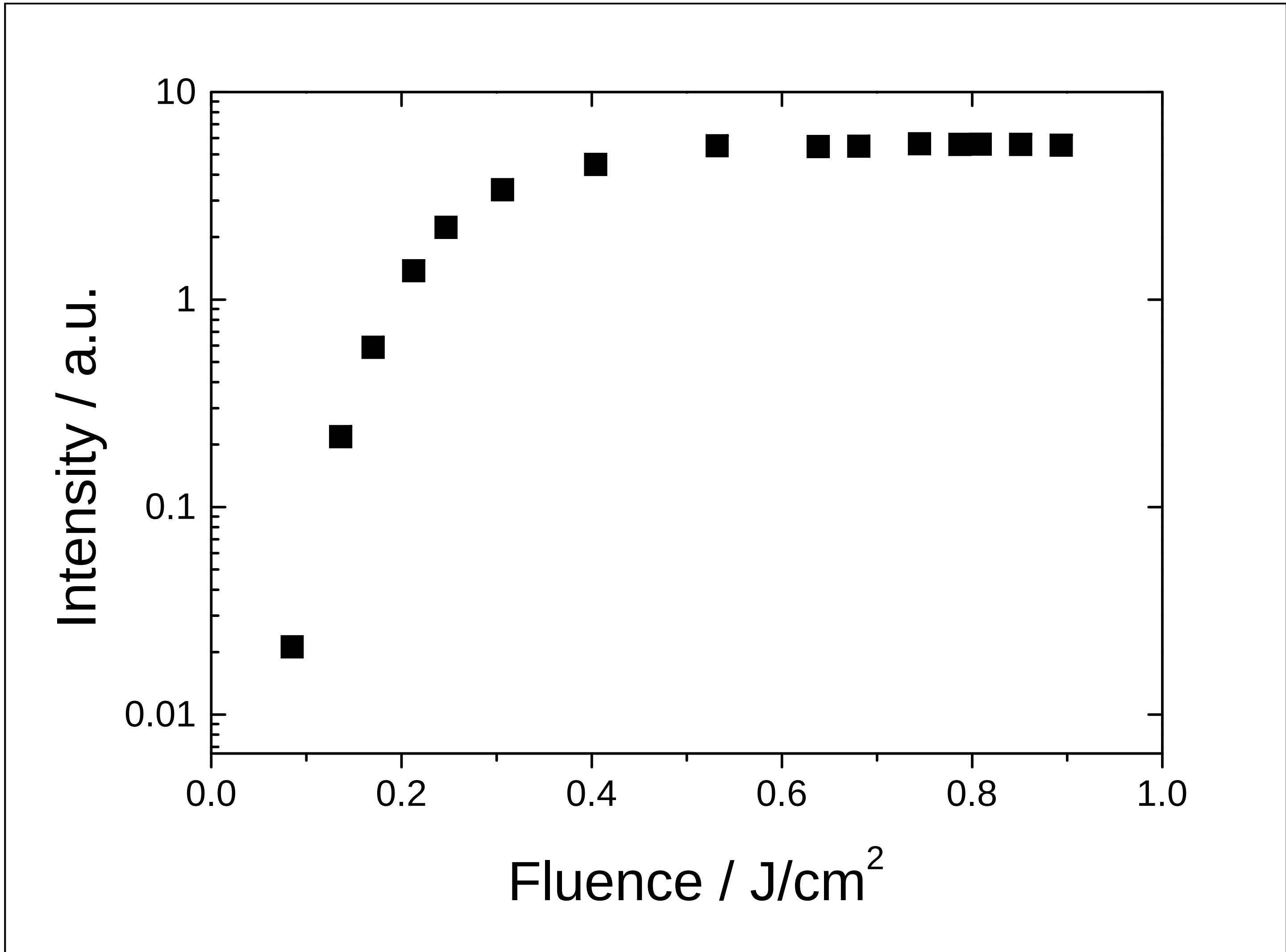
- Heat transfer
- Vaporization
- Emission of Planck radiation



The energy balance equation:

$$\underbrace{\frac{\pi^2 D^3 E(m)}{\lambda} q(t)}_{\text{Absorption}} - \underbrace{\frac{2k_a(T-T_g)\pi D^2}{D+G\lambda_{MFP}}}_{\text{HeatTransfer}} - \underbrace{\frac{\Delta H_v}{M_v}\frac{dM}{dt}}_{\text{Vaporization}} - \underbrace{\pi D^2 \int \varepsilon(D,\lambda) M_\lambda^b(T,\lambda)\, d\lambda}_{\text{PlanckRadiation}} - \underbrace{\frac{1}{6}\pi D^3 \rho_s c_s \frac{dT}{dt}}_{\text{InternalEnergyChange}} = 0$$

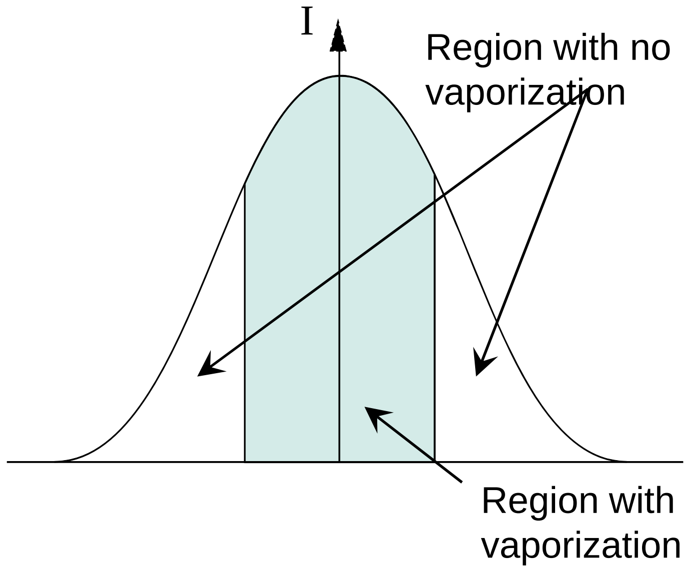
LII as a function of laser fluence

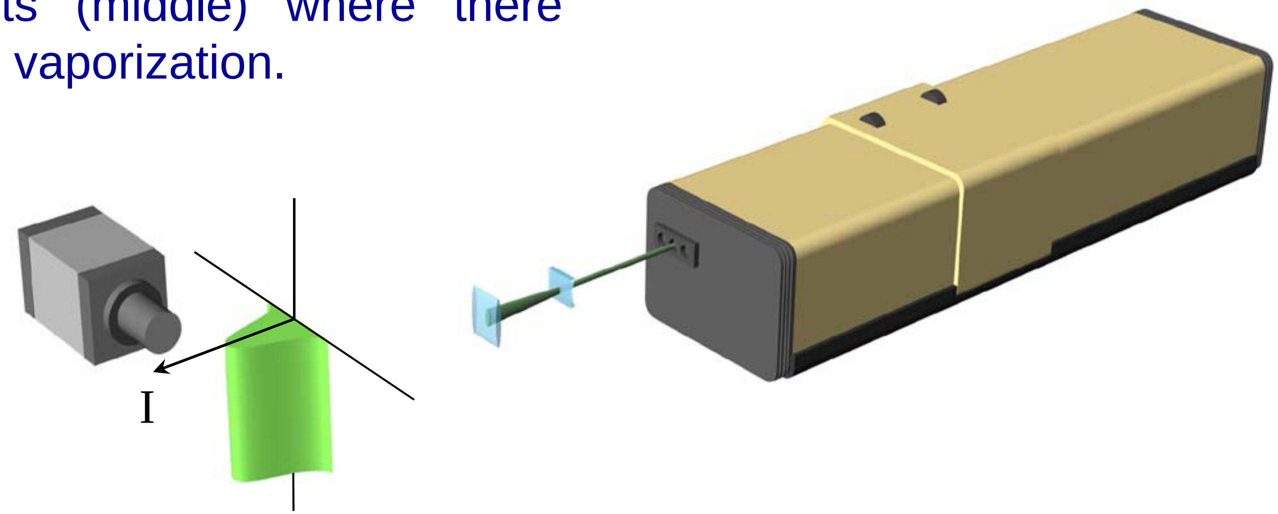


LII dependence on the laser spatial profile

The spatial profile of the laser drastically affects the LII signal.

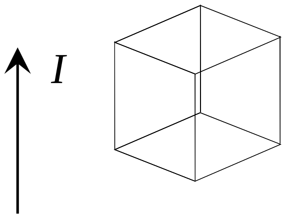
The laser illuminates particles with different energies. This means that contributions comes from both parts (edges) where there is no vaporization and parts (middle) where there are vaporization.



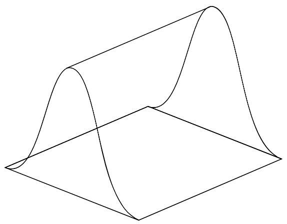


LII dependence on the laser spatial profile

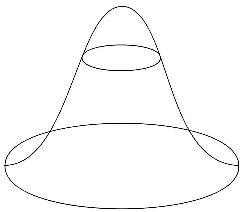
LII experimentalists often refer to the LII fluence curve. A fluence curve is the integrated LII signal plotted as a function of the laser fluence – that is the laser pulse energy divided by the exposure area of the laser beam. Below is shown modeled fluence curves for three different spatial profiles.



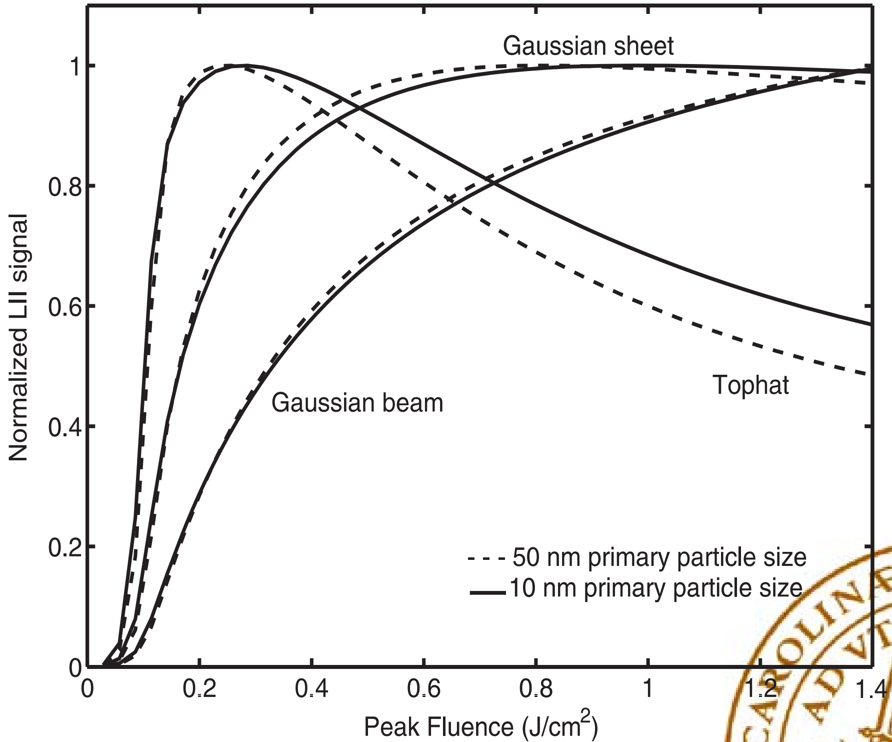
Tophat



Gaussian sheet



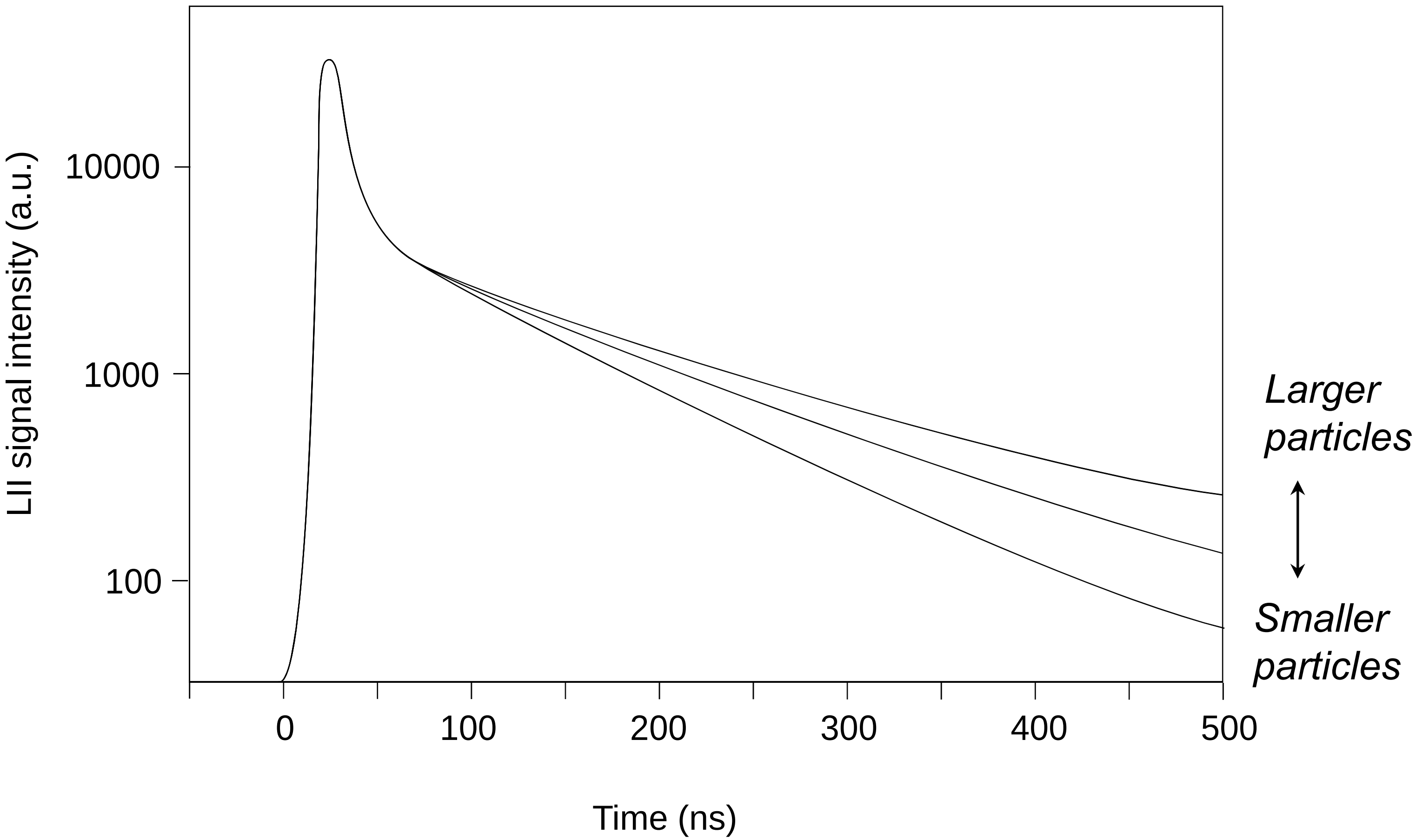
Gaussian beam



Calibration of LII by scattering/extinction

Time-resolved LII (TIRE-LII)

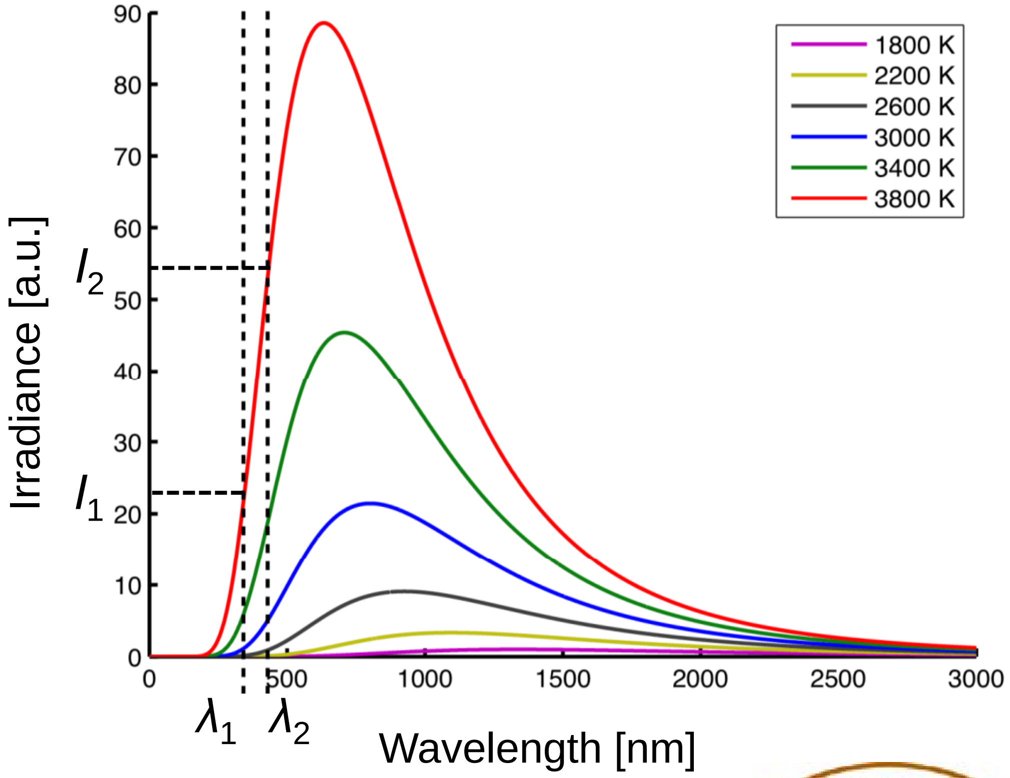
Detection of the time-resolved LII signal may yield the primary particle size. The decay time reflects the particle cooling and small particles cool faster than large ones!



Principle of 2-color LII

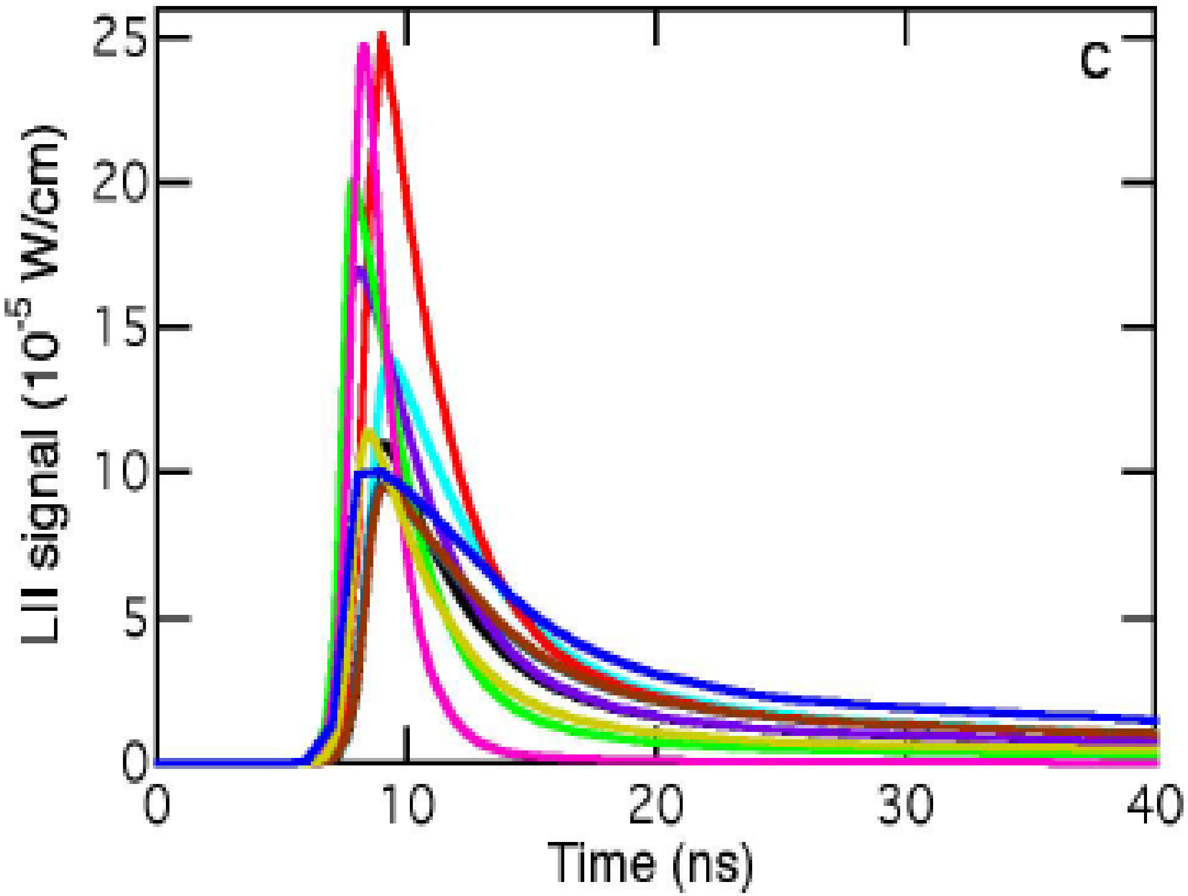
- Used to minimize uncertainties in evaluated particle sizes from LII signal decays as the rate of absorbed laser energy does not need to be known.
- The temperature is calculated using the relative irradiance at two wavelengths.

$$T = \frac{hc}{k} \cdot \frac{1/\lambda_2 - 1/\lambda_1}{\ln\left(\frac{I_1}{I_2} \cdot \left(\frac{\lambda_1}{\lambda_2}\right)^6\right)}$$



Courtesy: Henrik Bladh

Modelling of time-resolved LII signals



Comparison of LII models between different research groups

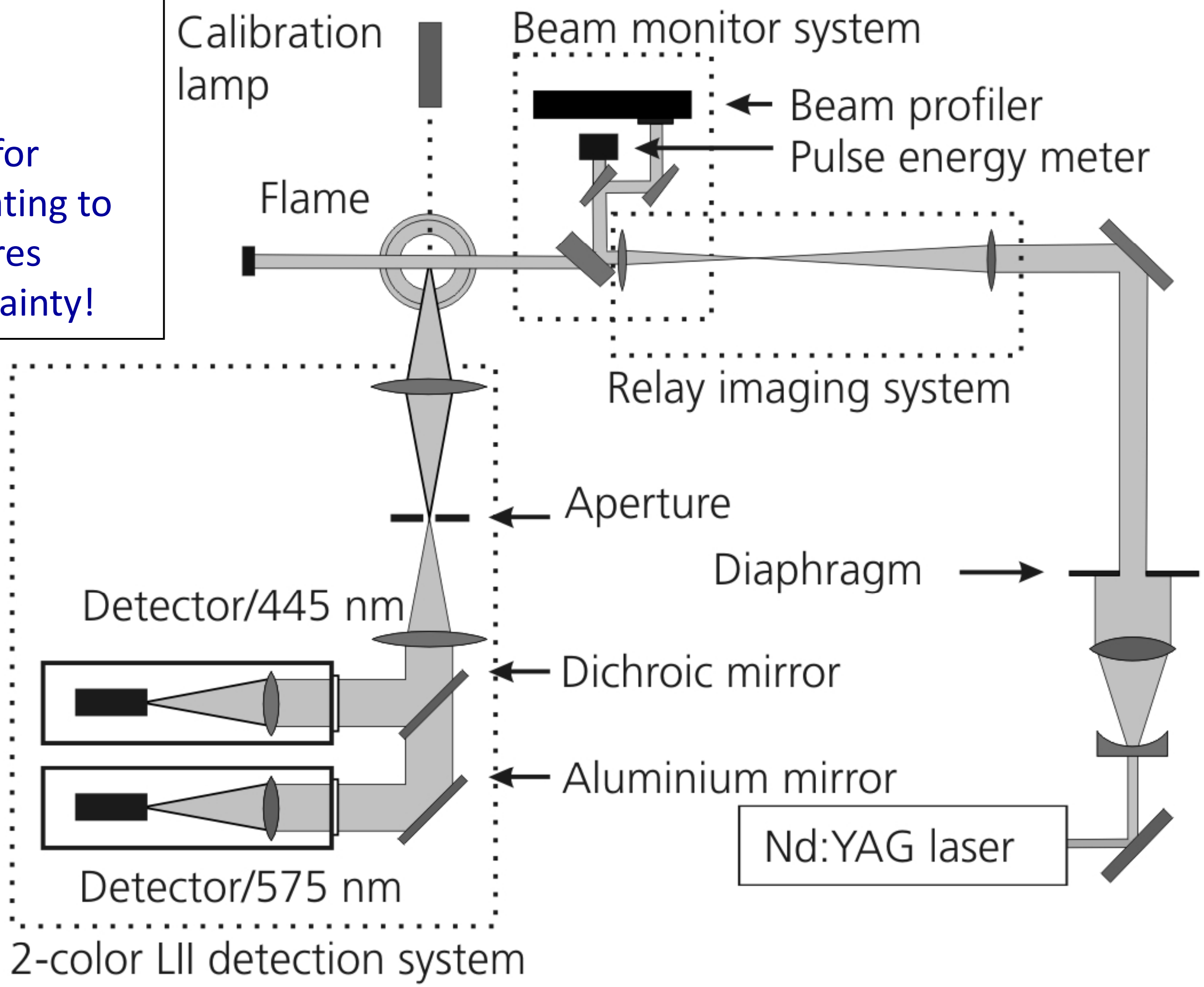
- Same experimental input
- Unconstrained models
- High fluence (0.7 J/cm^2)

Michelsen et al., Modelling of laser-induced incandescence of soot: A summary and comparison of LII models, submitted to Applied Physics B

Experimental setup for two-color LII

Tophat spatial laser profile
0.13 J/cm^2
1064 nm

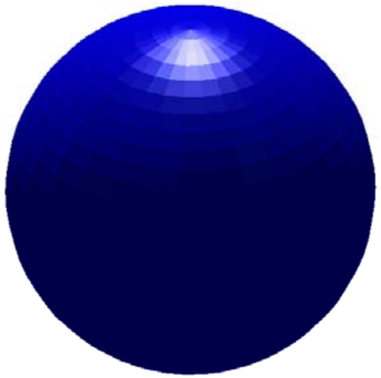
Tophat profile used for particle sizing as heating to different temperatures increases the uncertainty!



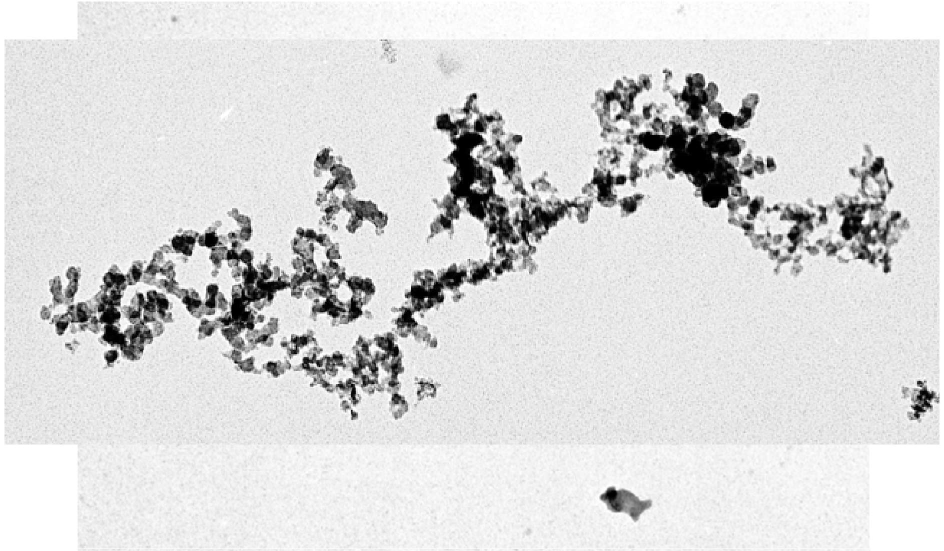
From: Bladh, H., et al., *Proc. Combust. Inst.* ***33,*** *641-648 (2011)*

Modelling a soot particle in LII

Typical model



Real-world examples (microscopy)

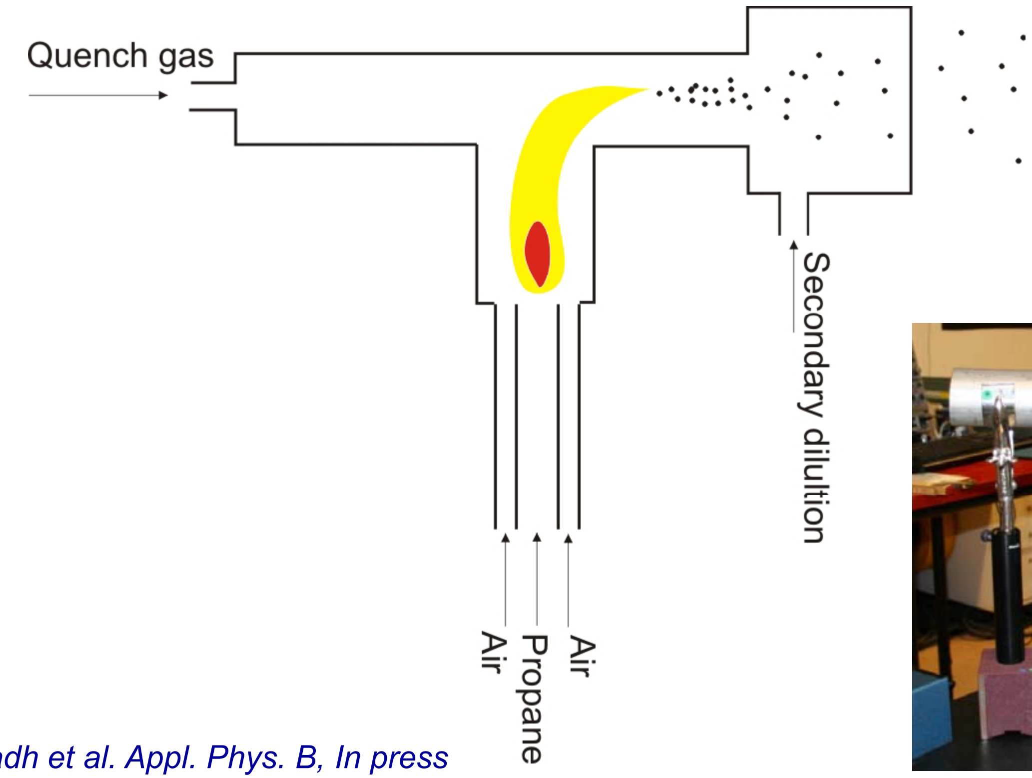


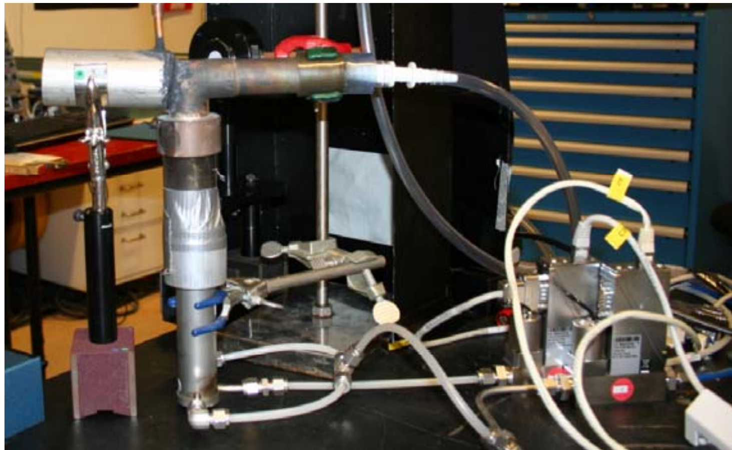
Shape described by:

- Diameter (*D*)
- Primary particle diameter (D)
- Number of primary particles
- Radius of gyration of aggregate
- Fractal parameters (”compactness”)
- etc.
- No particle looks the same as any other!

The effect of aggregation on LII signals

- Theory shows that LII signals should be affected by the level of soot aggregation, i.e. decay rate/shape dependent on both primary particle and aggregate size!
- First experimental evidence for this effect using LII on a cold soot source: Soot generator based on a quenched diffusion flame

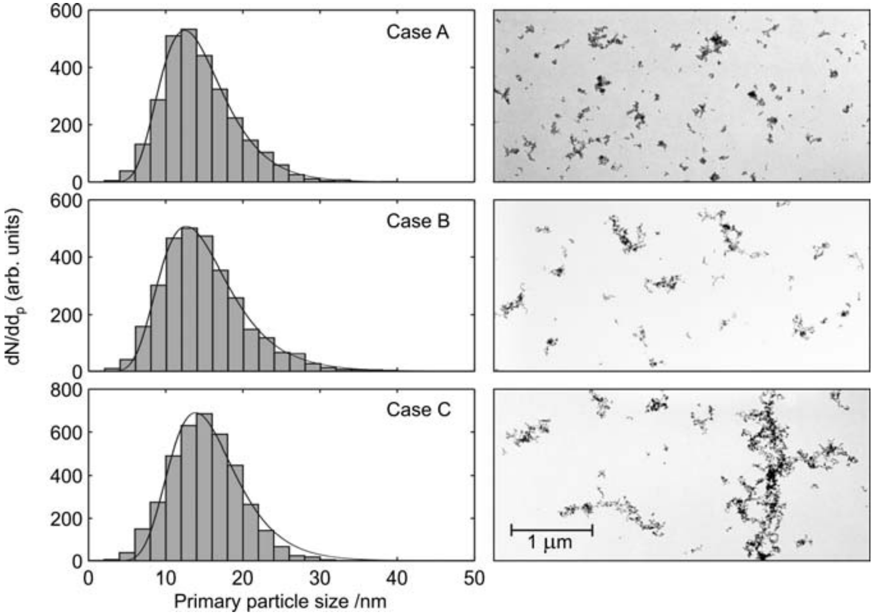




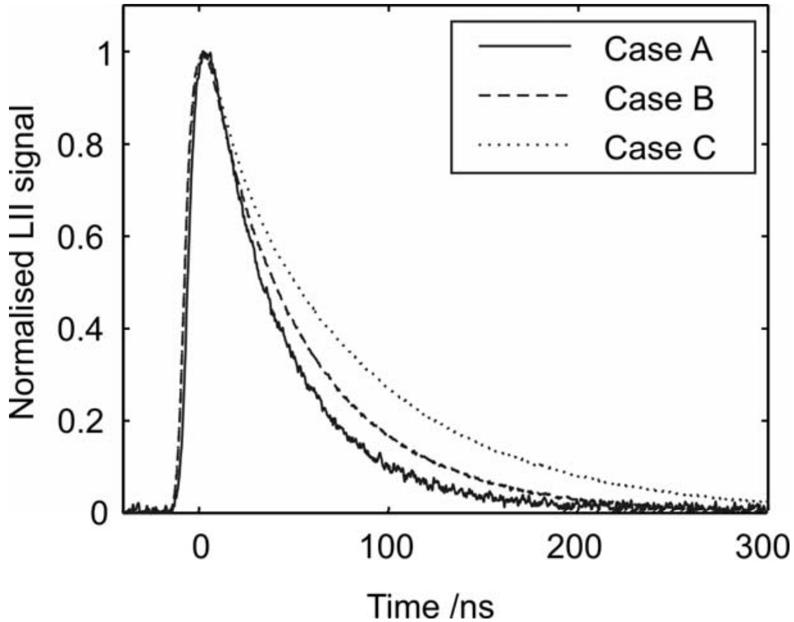
Bladh et al. Appl. Phys. B, In press

The effect of aggregation on LII signals

- First experimental evidence for this effect:



TEM data on primary particle sizes: Similar primary particle sizes!

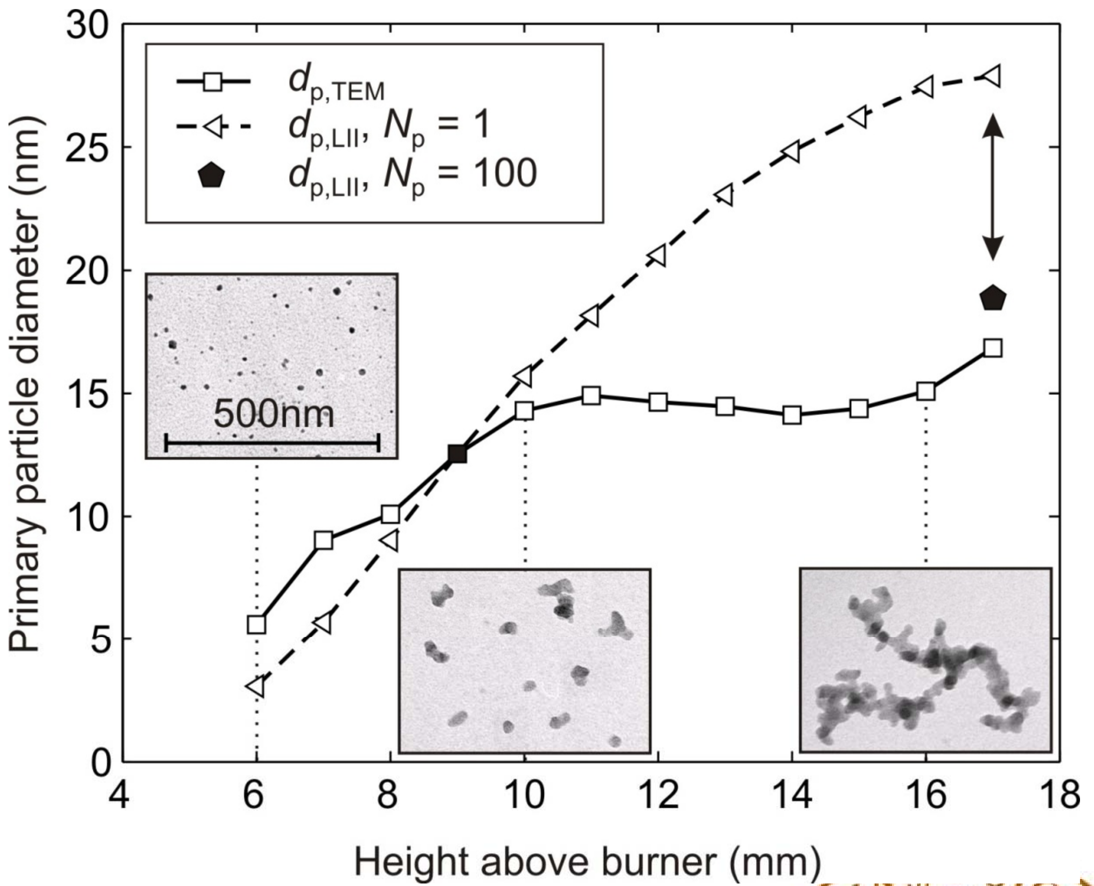


LII signals: Different decay rates

Bladh et al. Appl. Phys. B, In press

The effect of aggregation on LII signals

- Sampling using a pneumatic probe
- Large differences between TEM and LII sizes
  - Deviation above 10 mm HAB may be due to aggregation
  - Uncertainties in TEM sampling procedure and analysis

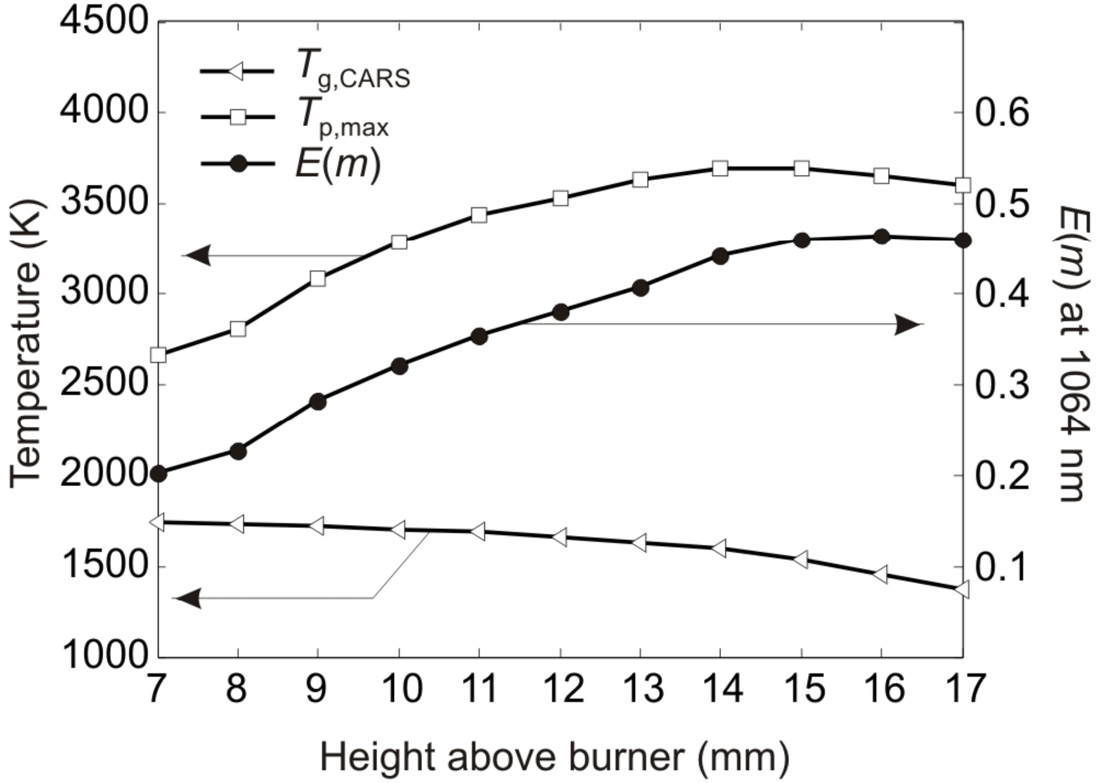


⬟ Aggregation model from Liu et al. 2006, Appl. Phys. B, 83, 383-395

Bladh, H., et al., *Proc. Combust. Inst.* ***33,*** *641-648 (2011)*

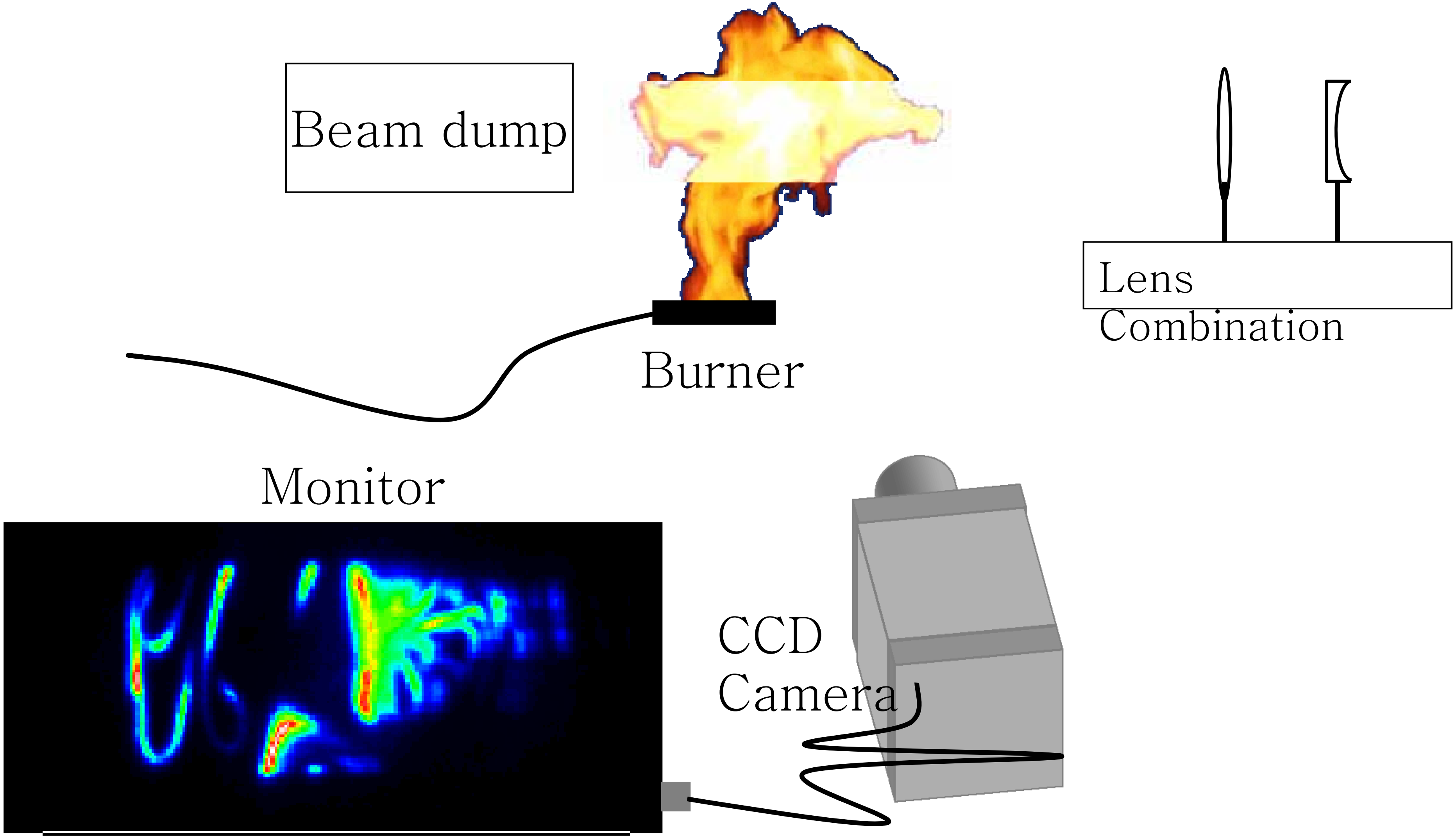
E(*m*) as function of height above burner (HAB)

- Evaluate difference between gas temperature and maximum soot temperature for each height!
  - Gas temperature from rotational CARS
  - Maximum temperature from two-color pyrometry
- $$E(m) = -\mathrm{Im}\left\{\frac{m^2-1}{m^2+2}\right\}$$
- Procedure as described by Snelling et al. 2004, Combust. Flame, 136, 180-190
- LII model to determine *E*(*m*)

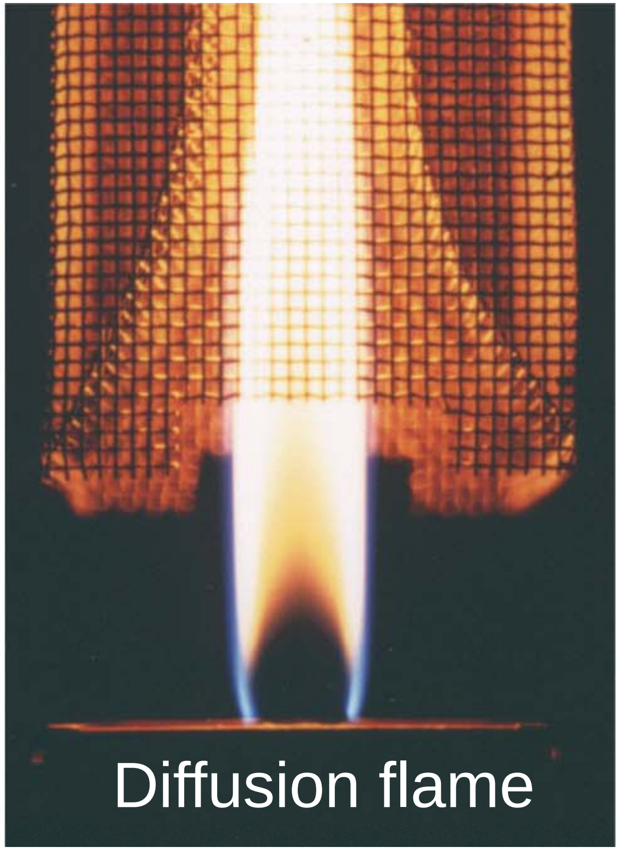


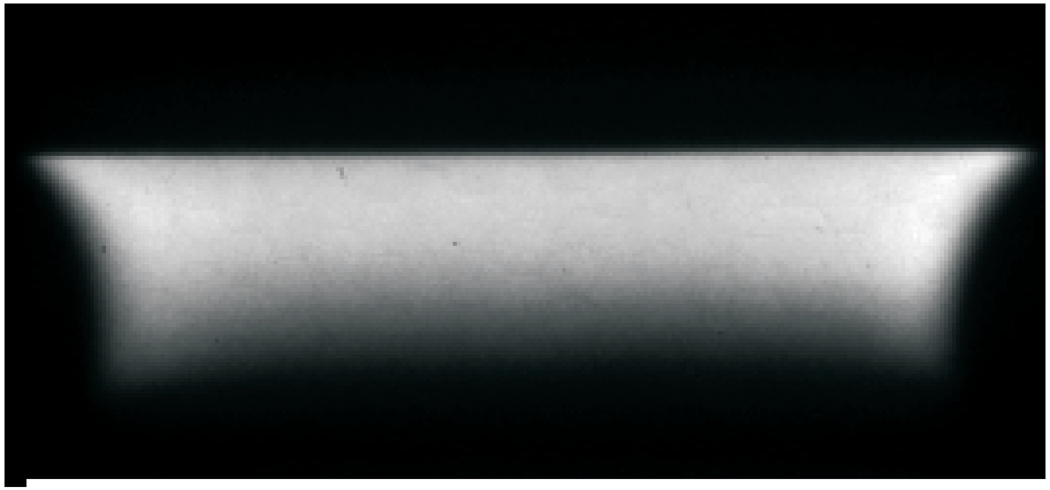
Bladh, H., et al., *Proc. Combust. Inst.* ***33,*** *641-648 (2011)*

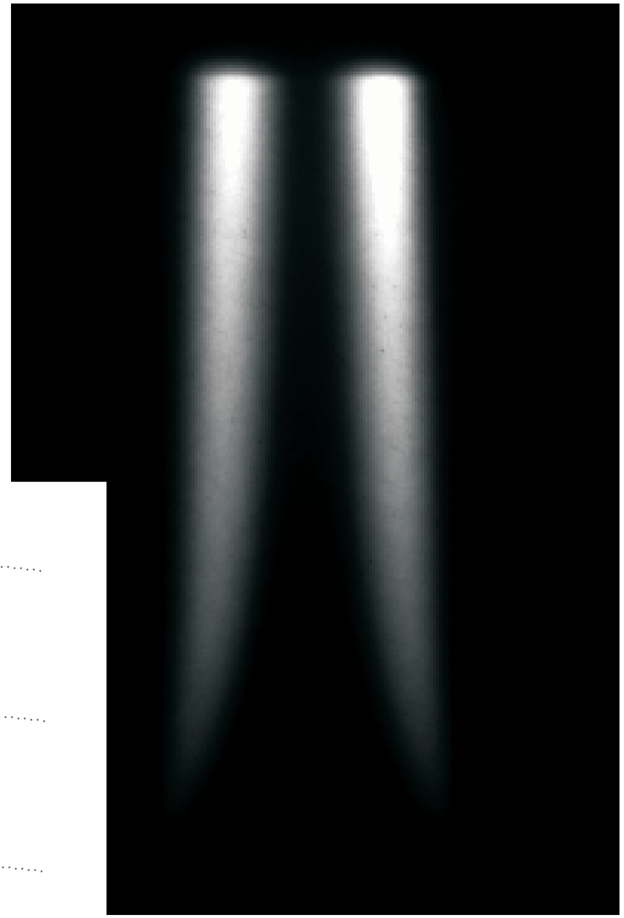
LII 2D measurements

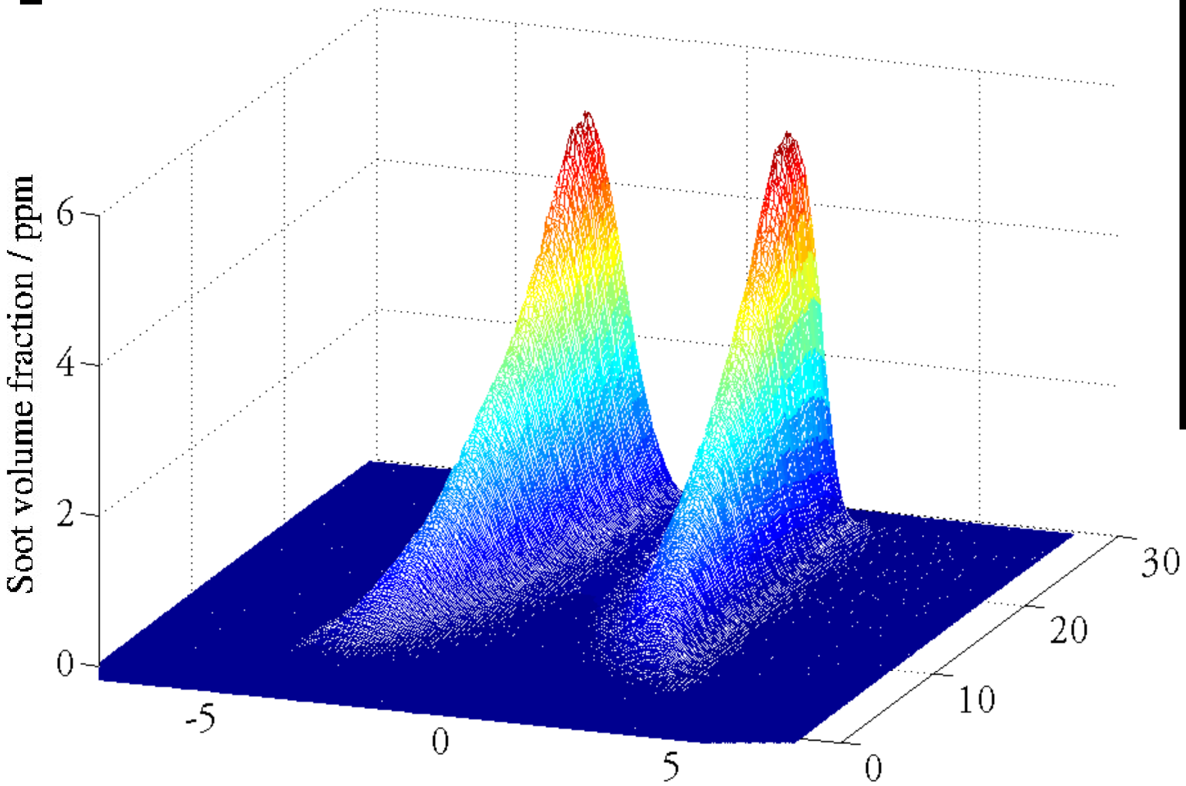



Premixed flame

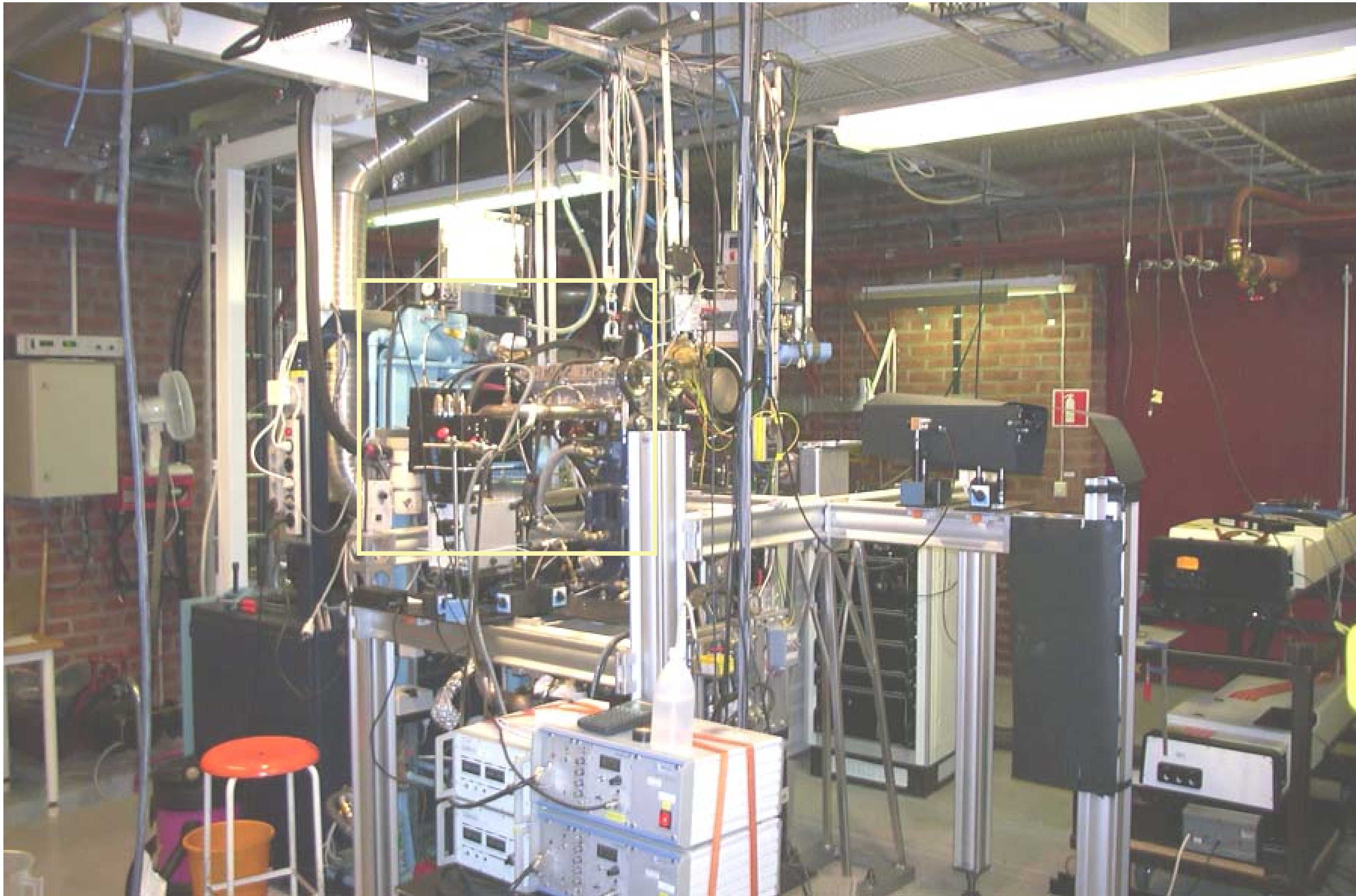

Diffusion flame

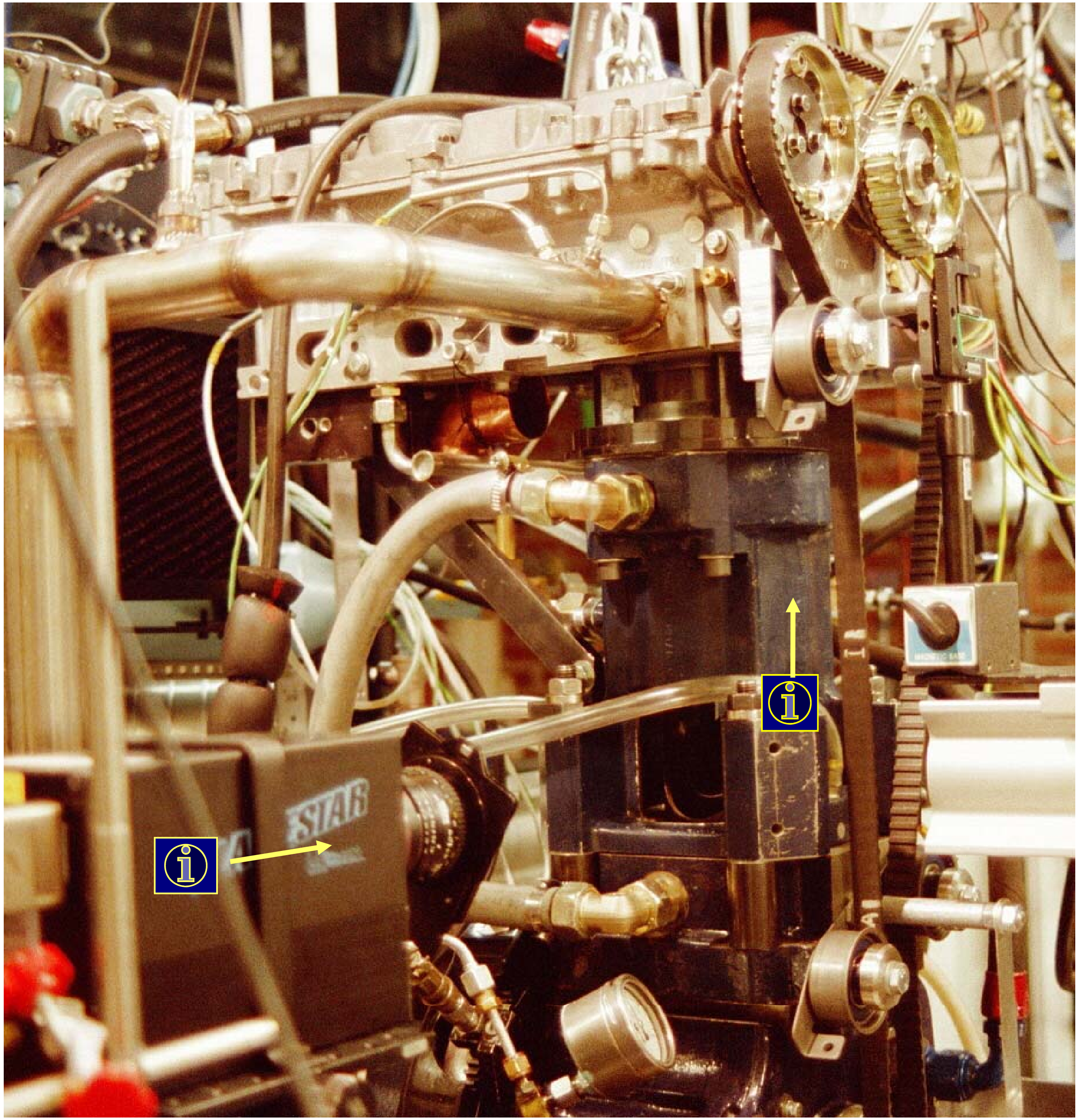




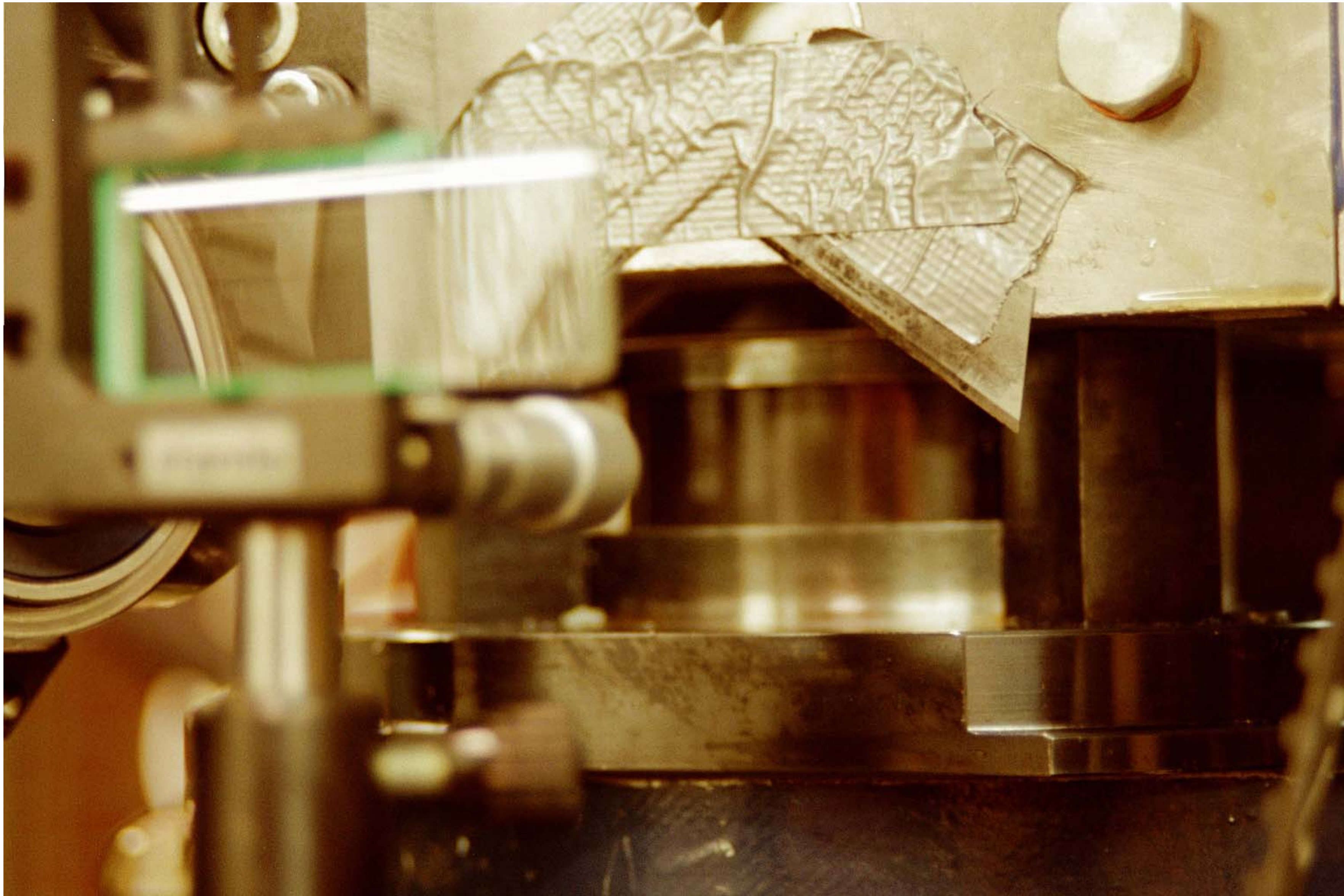

Soot volume fraction / ppm
0
2
4
6
-5
0
5
0
10
20
30

Engine Measurements



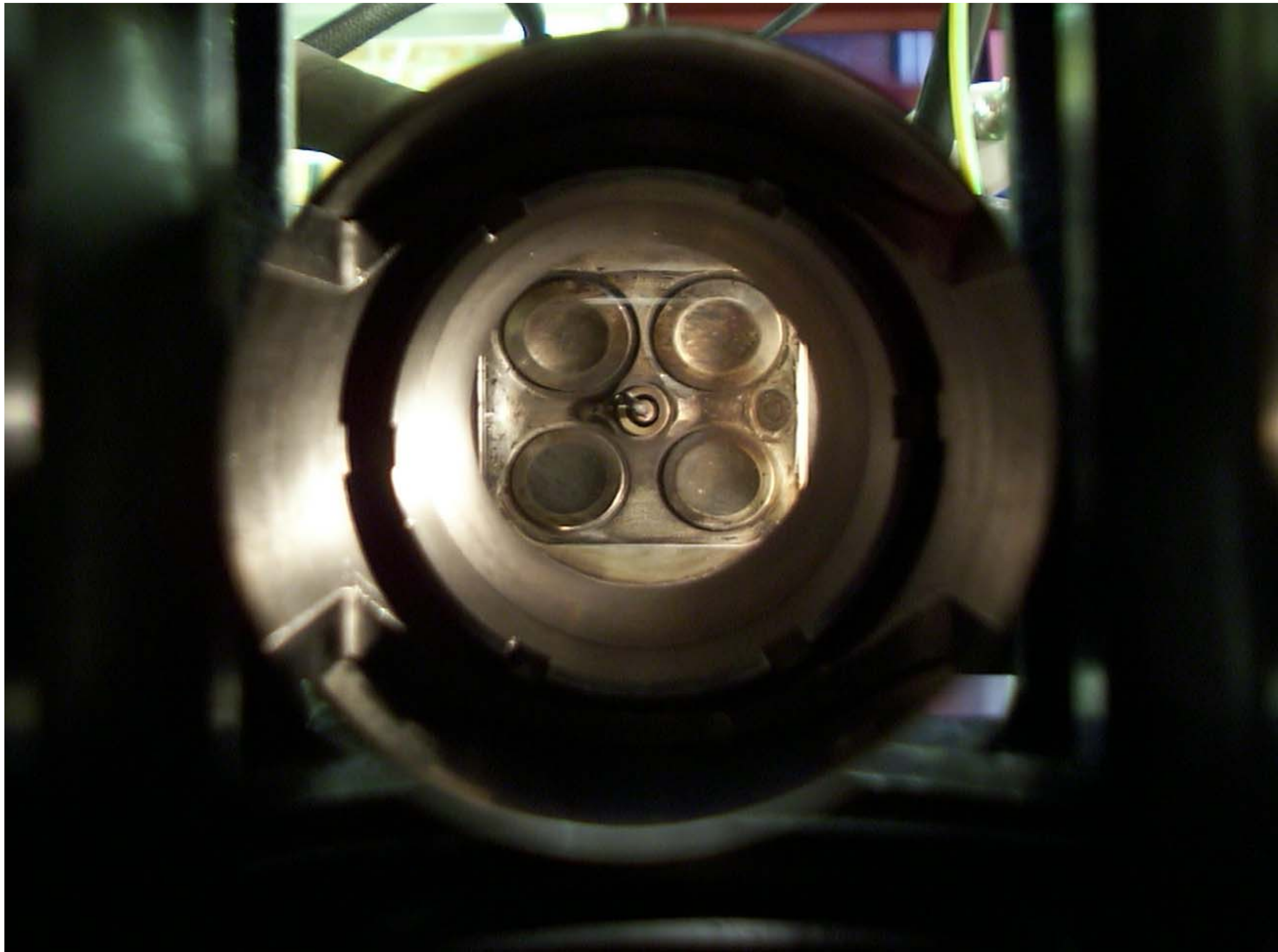

STAR

Laser entrance to the engine



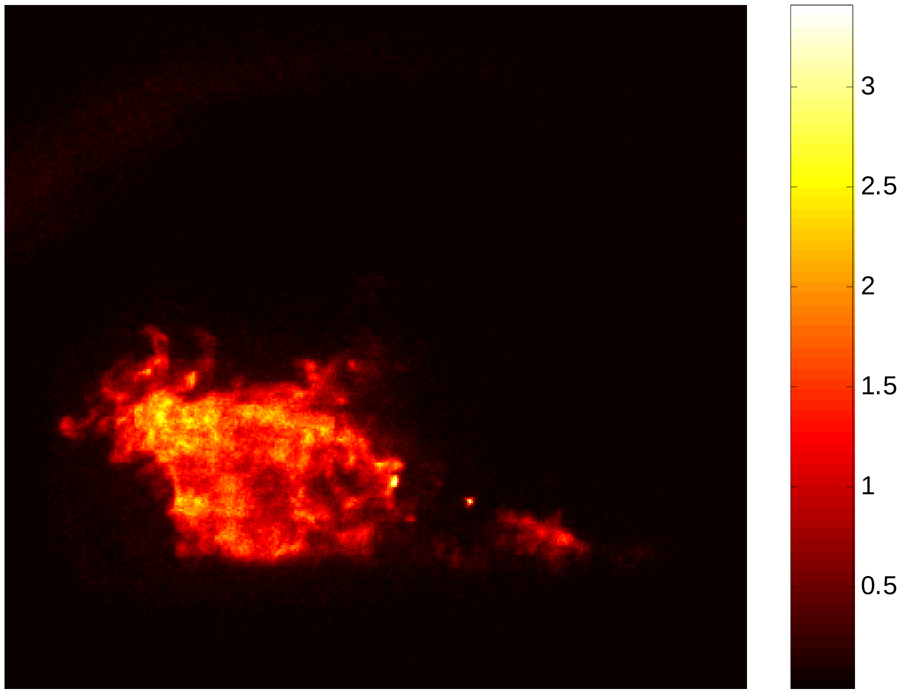
Investigation though the piston

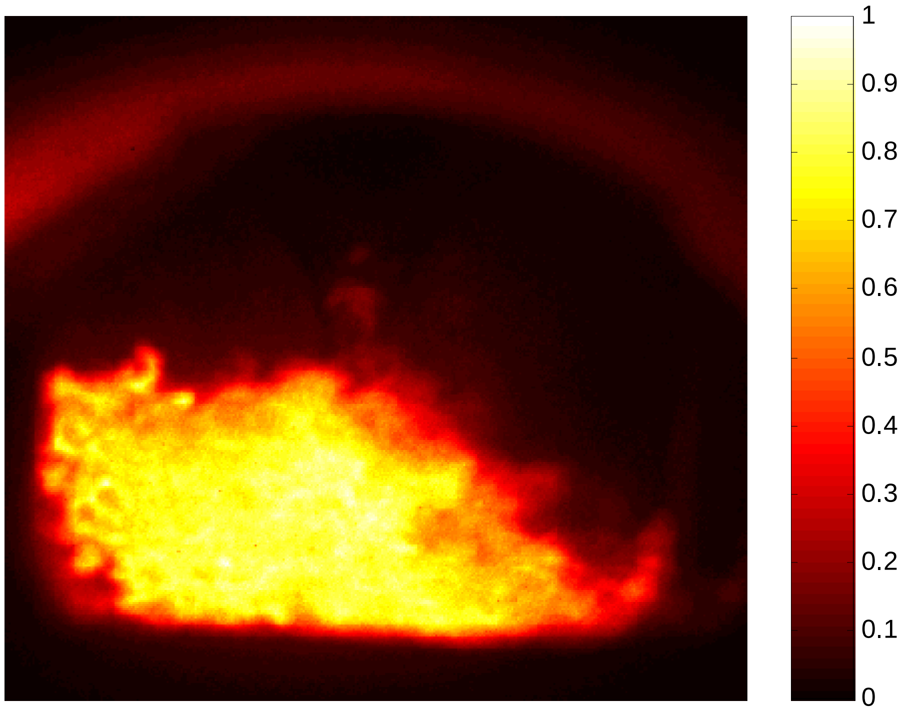
Investigation though the piston



LII signal

Flame luminosity

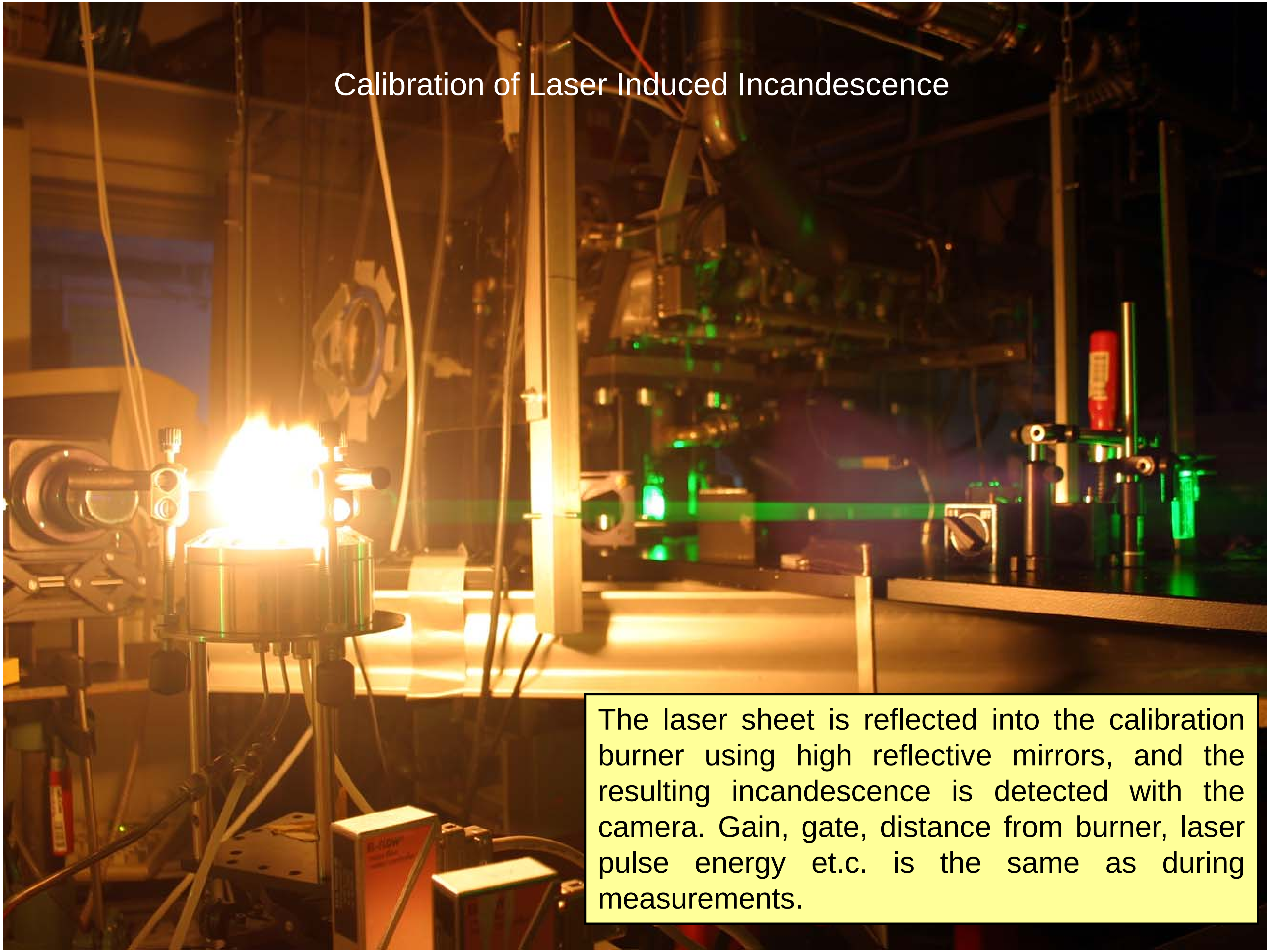




Detection from below, through the piston

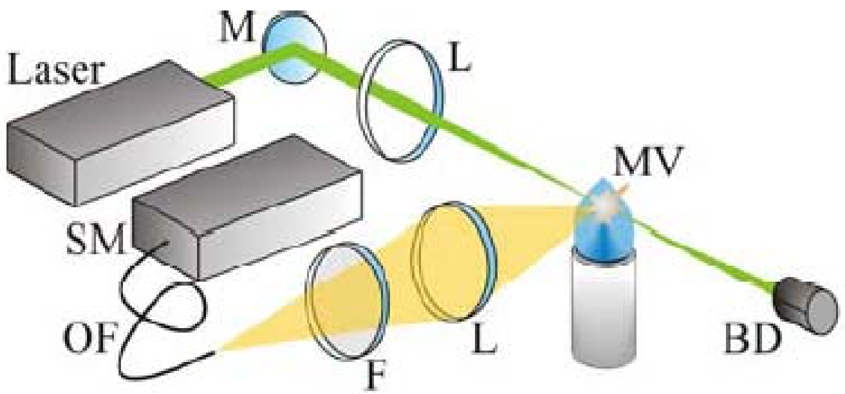
Left image shows soot volume fraction in parts per million, ppm.

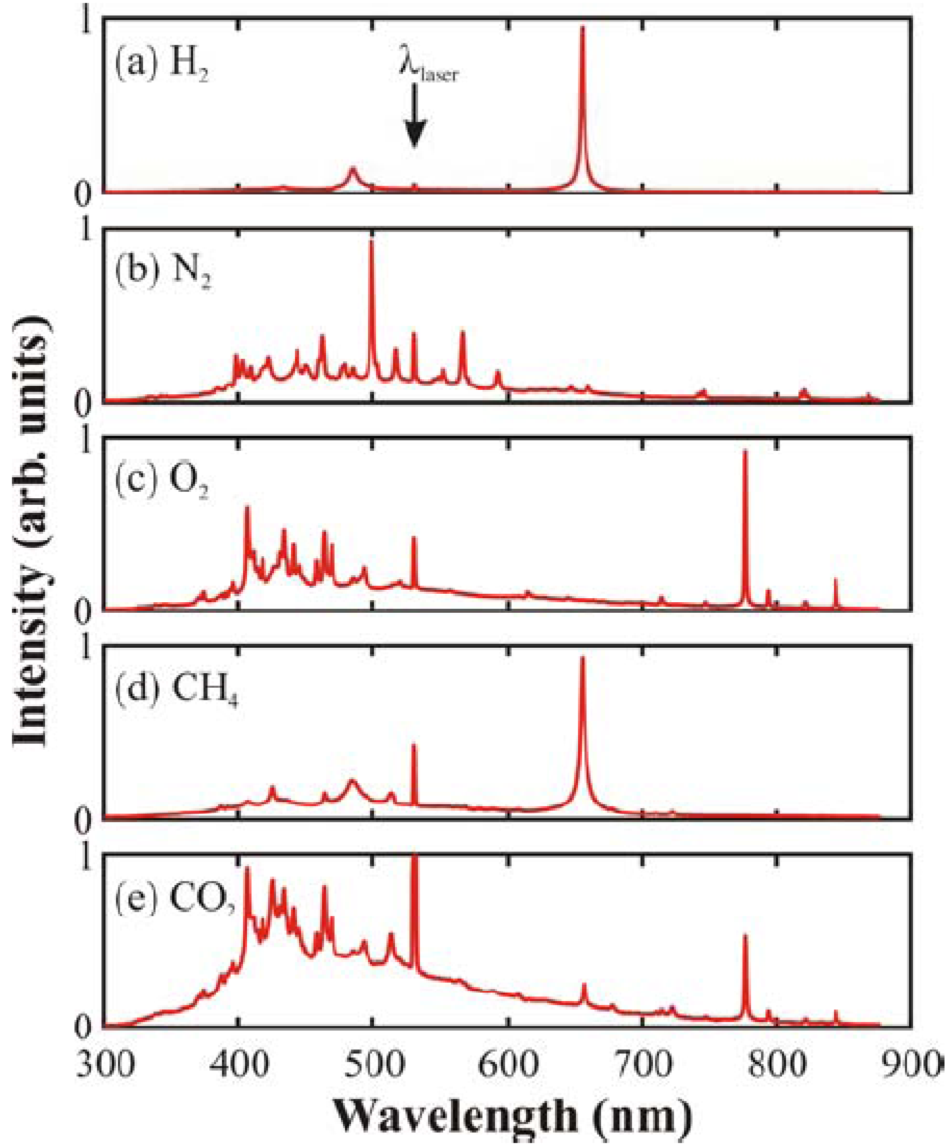
Calibration of Laser Induced Incandescence



The laser sheet is reflected into the calibration burner using high reflective mirrors, and the resulting incandescence is detected with the camera. Gain, gate, distance from burner, laser pulse energy et.c. is the same as during measurements.

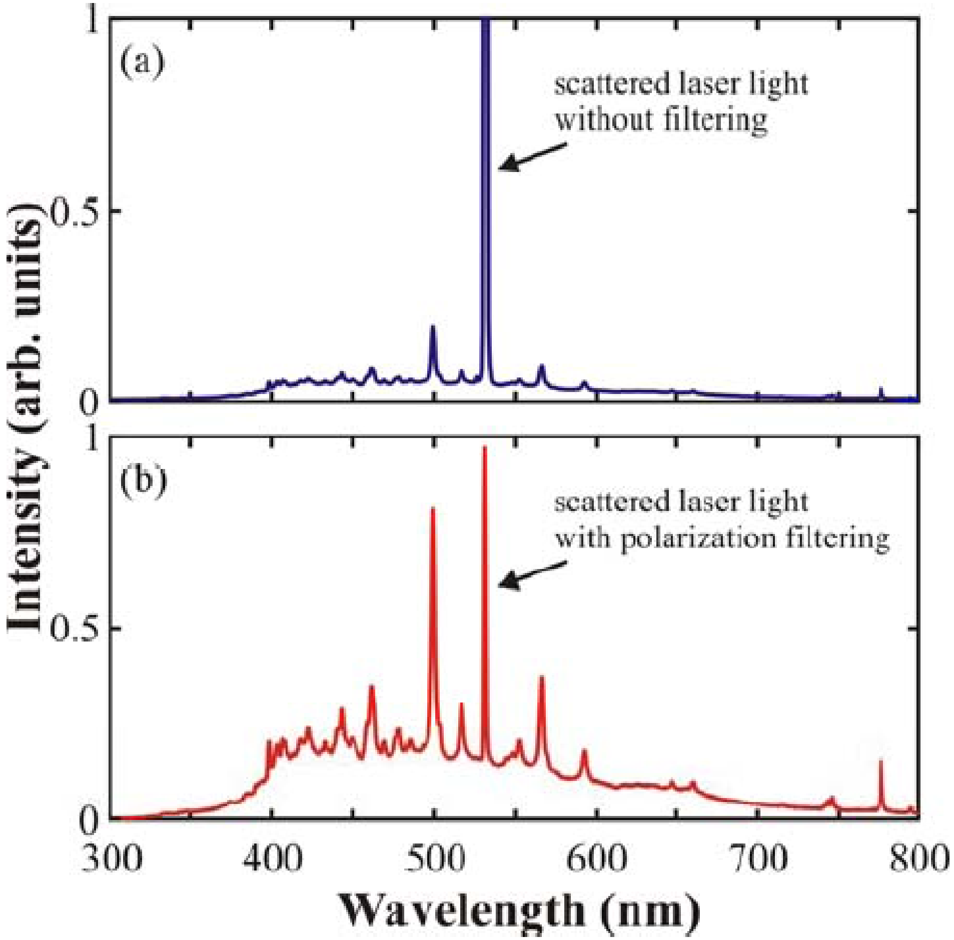
II. Laser-induced breakdown spectroscopy - LIBS

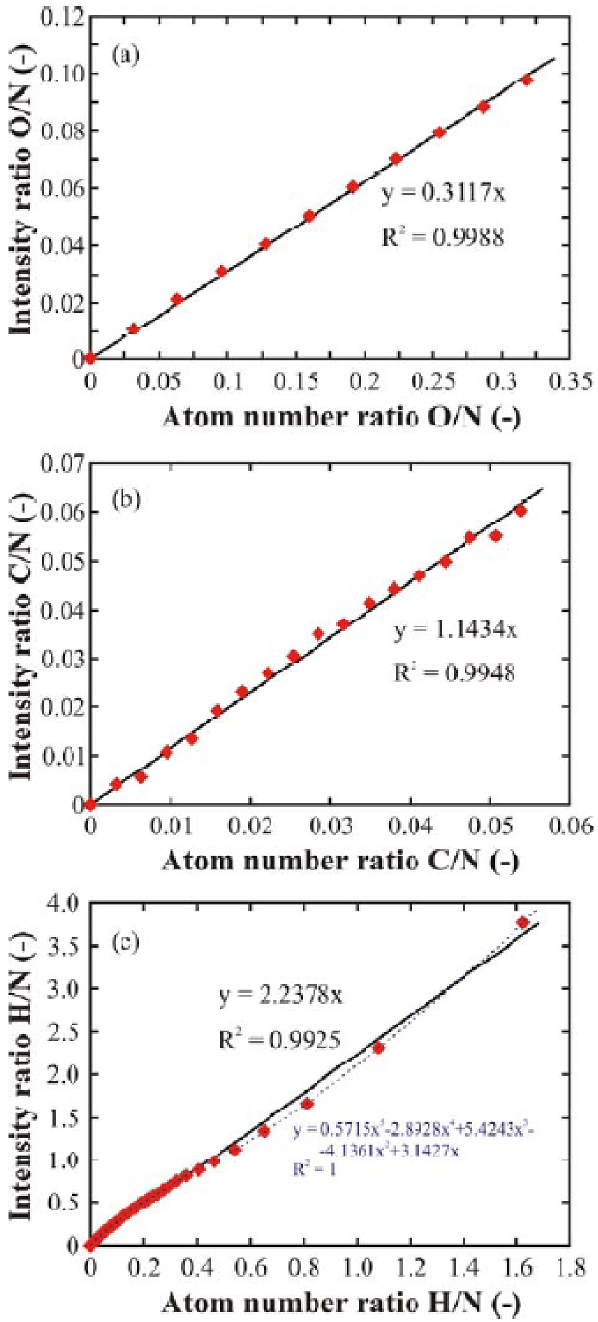




J. Kiefer, J.W. Tröger, T. Seeger, A. Leipertz, B. Li, Z.S. Li and M. Aldén, 'Laser-induced breakdown spectroscopy in gases using ungated detection in combination with polarization filtering and online background correction', Measurement science and technology 21, 065303 (2010).

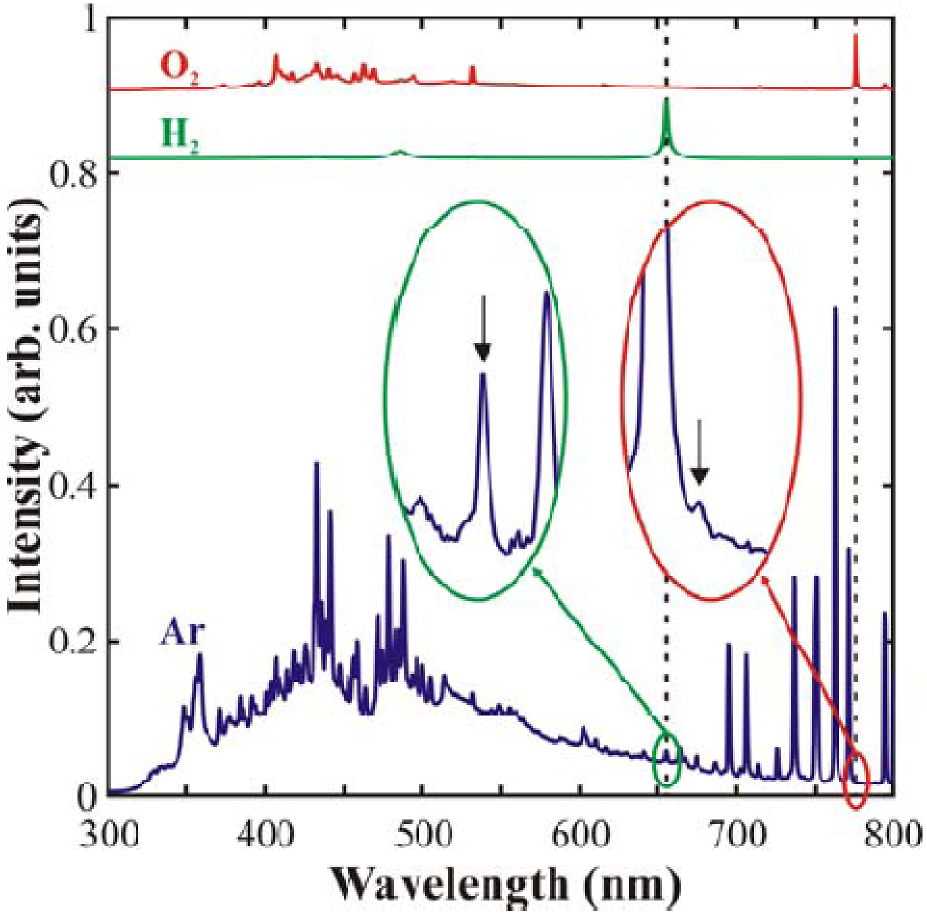
Polarization suppression of laser light - calibration





Calibration curves

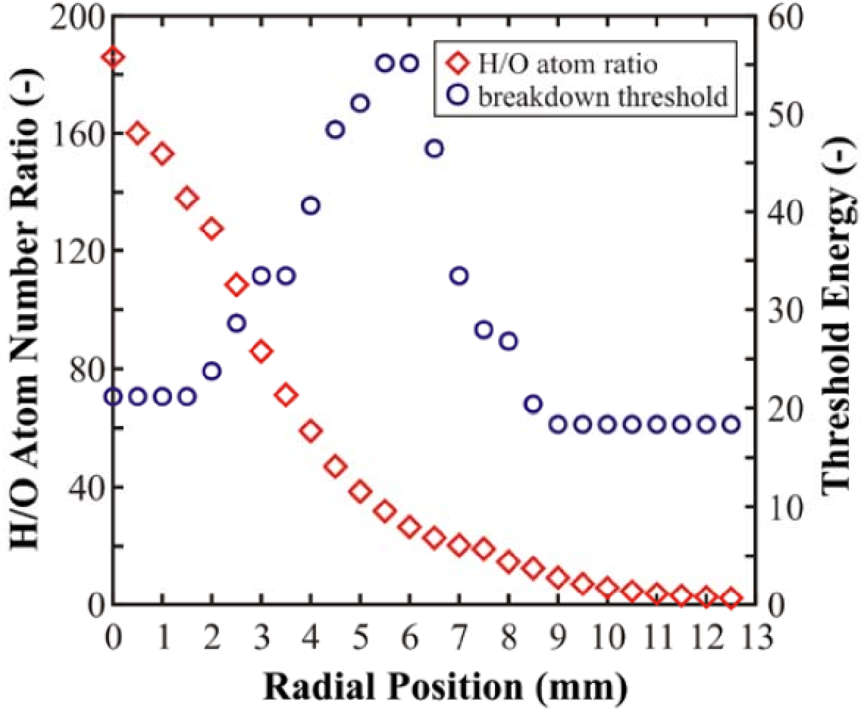
Detection limits



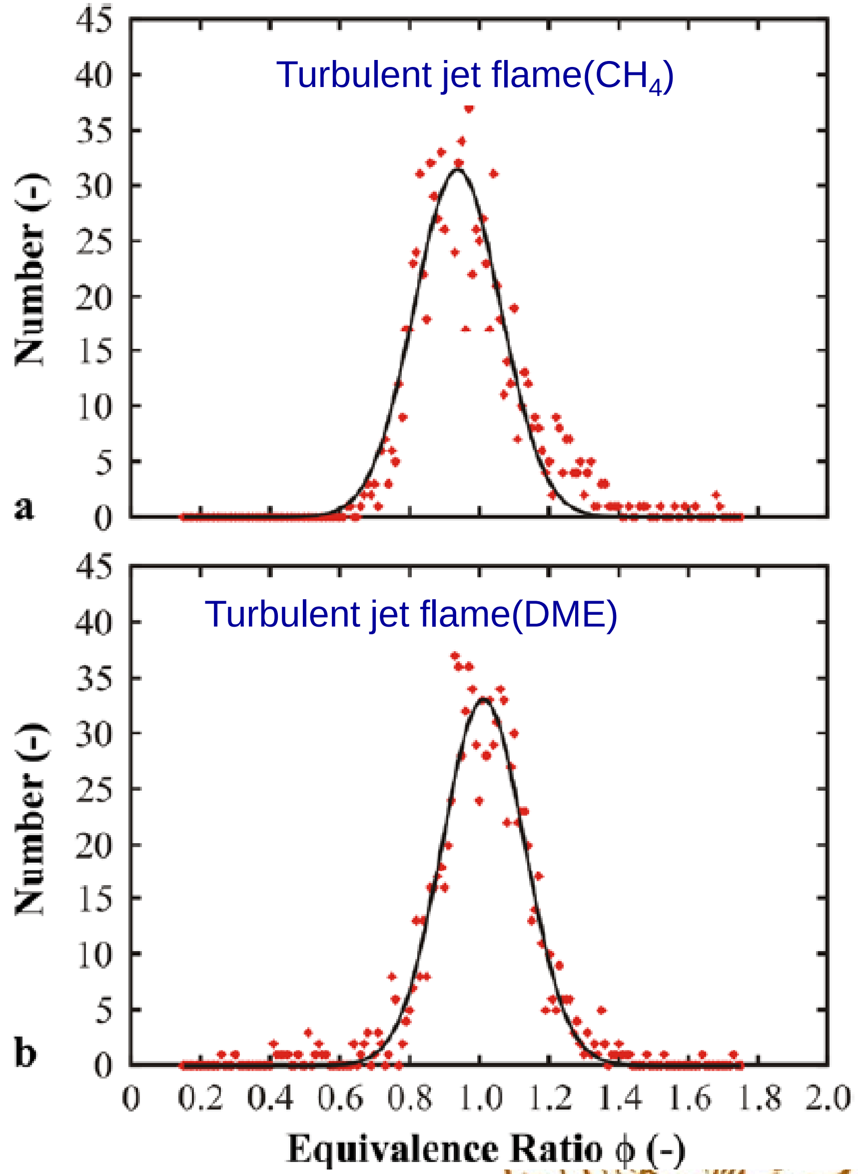
Argon gas with a nominal purity of 99.996% with small contents of carbon dioxide (<1 ppm), oxygen (<4 ppm), water (<5 ppm) and nitrogen (<10 ppm).

J. Kiefer, J.W. Tröger, T. Seeger, A. Leipertz, B. Li, Z.S. Li and M. Aldén, 'Laser-induced breakdown spectroscopy in gases using ungated detection in combination with polarization filtering and online background correction', Measurement science and technology 21, 065303 (2010).

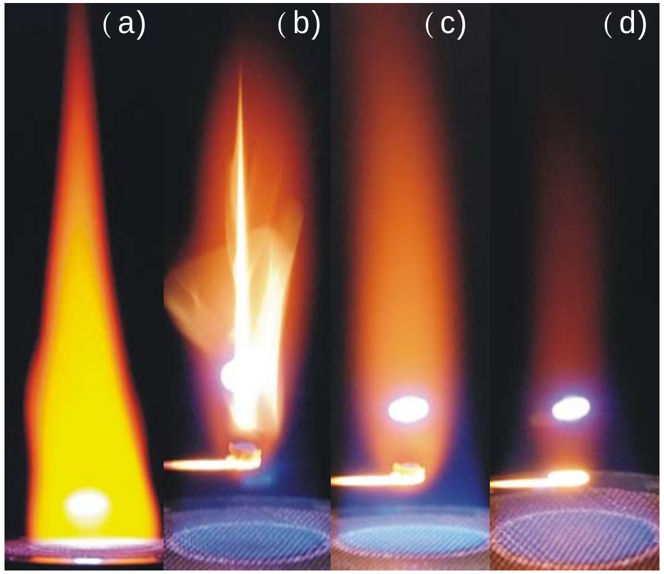
Application I: Flames

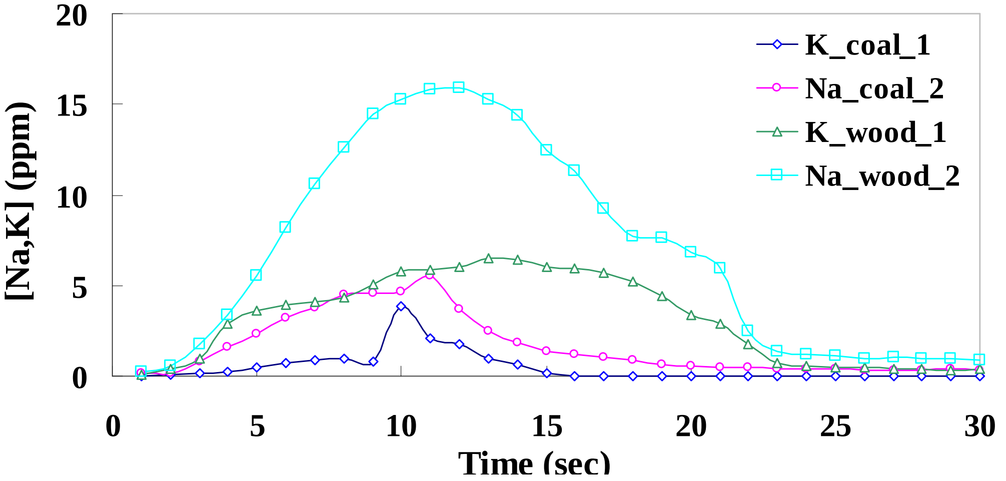


Atom ratio H/O and breakdown threshold as a function of radial position in a non-premixed methane flame



Application II: Measurements of Na and K from coal/biomass





LIBS is operated in (a) seeded flame (b) devolatilization (c) char (d) ash cooking phases

Release of sodium and potassium during devolatilization with equivalence ratio of 1.3 using LIBS.