Laser-induced incandescence techniques for soot measurements: capabilities, opportunities, and challenges

Liu, Fengshan

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Laser-Induced Incandescence Techniques for Soot Measurements: Capabilities, Opportunities and Challenges

Fengshan Liu

Institute for Chemical Process and Environmental Technology
National Research Council Canada
Ottawa, ON, Canada K1A 0R6

中国工程热物理学会 **2009** 年燃烧学学术年会
31 October 2009, Hefei, China

National Research Council Canada Conseil national de recherches Canada

Canada

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Outline

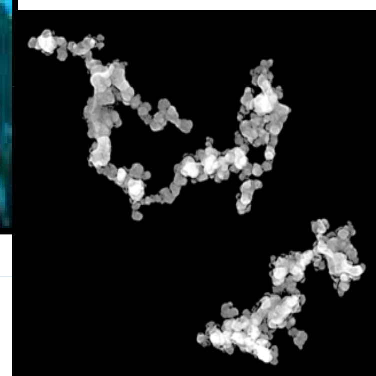
- Background
- A brief history of LII
- Introduction of LII: principle, setup, issues
- LII Theory
- LII applications in combustion: capabilities and opportunities
- Remaining challenges
- Conclusions

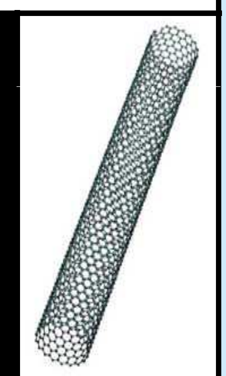
Background

Desired and undesired aspects of soot

Improve efficiency of combustion technology

Engineered nanoparticle synthesis





Assess the ecological and health impacts of combustion

Background

- Emission of soot from various combustion devices (automobiles, furnaces, gas turbines) into the environment is detrimental to human health and a major contributor to climate change
- Our understanding of soot formation is still quite limited
- Modern and future generation combustion devices (engines, combustors) produce less soot, which requires highly sensitive diagnostic techniques
- Morphological information of soot (fractal properties, primary particle size, aggregate size distribution) is required to gain further fundamental insights into soot formation and to assess their health impact

Background

- It is desirable to develop real-time, in-situ, and sensitive measurement techniques to meet these challenges
- Conventional techniques (e.g., light extinction, laser scattering) do not meet these requirements
- Laser-Induced Incandescence (LII) is a promising candidate technique and has been rapidly developed into a powerful diagnostic tool for soot and other nanoparticle characterization

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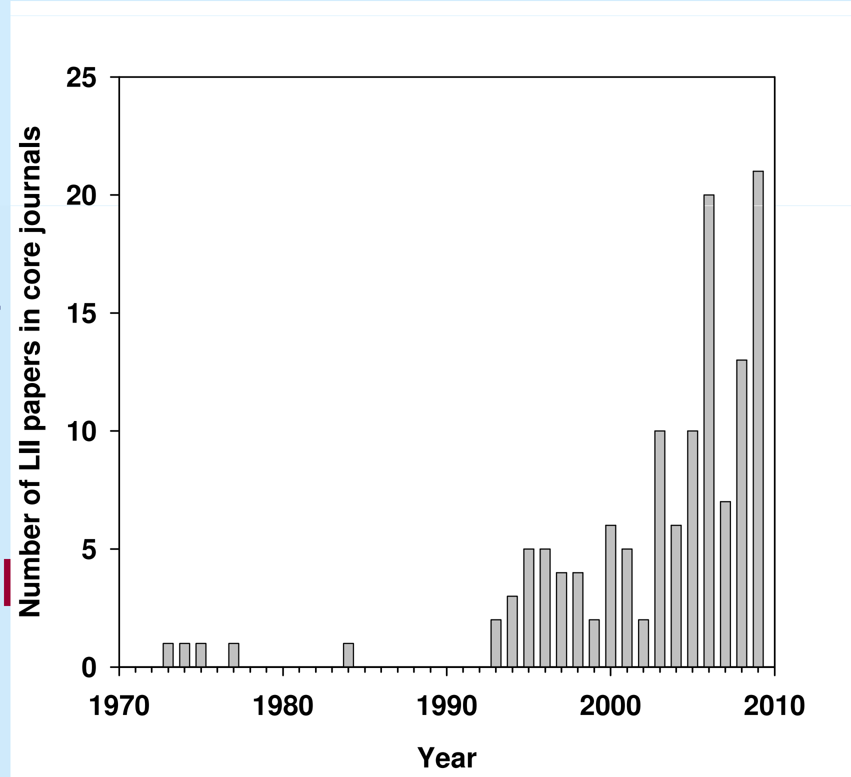
History of LII

- The first reported study related to LII was perhaps made by Gelbwachs and Brinbaum in **1973** (Applied Optics) (Aerospace Corp., California)
- They reported the phenomenon of broadband “fluorescence of atmospheric aerosols” as an interfering signal to the molecular fluorescence used for detecting gaseous pollutants (NO_2 in their case) using a cw argon ion laser (458 nm to 515 nm)
- They suggested that “Fluorescence is a potentially useful means for aerosol identification and monitoring.”
- In **1974**, Weeks and Duley (York University, Toronto) published the very first paper (Aerosol-particle sizes from light emission during excitation by TEA CO_2 laser pulses) to use the concept of LII as a means to determine particle size
- These researchers are the pioneers of LII by recognizing that “**the passage of intense laser radiation through a dusty atmosphere results in the momentary emission of visible light from particles in the path of the beam**.”

History of LII

- Because the two pioneering studies are not in the area of soot and flame, the LII community and literature paid no attention to these two important pieces of work
- **Eckbreth** in **1977** also observed the phenomenon of LII in a sooting laminar propane diffusion flame (JAP: Effects of laser-modulated particulate incandescence on Raman scattering diagnostics)
- The particle laser energy absorption and the soot particle evaporation processes were analyzed
- **Eckbreth** also detected LII signals at **two wavelengths** to monitor the peak soot particle temperature

History of LII



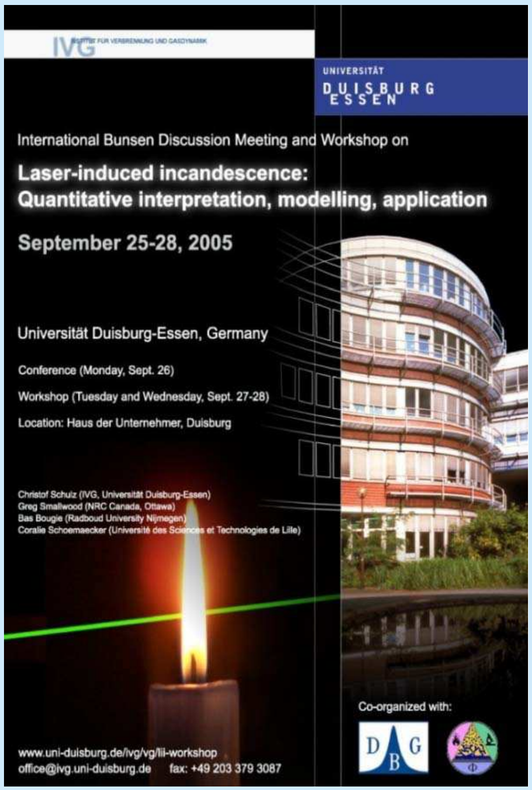
- Melton in **1984** published the first paper to explicitly explore LII as a diagnostic for soot measurement (Soot diagnostics based on laser heating)
- Melton also formulated the first LII model, which remains the backbone of subsequent models
- **Eckbreth**'s work and **Melton**'s were regarded as the birth of modern LII techniques for measurement of soot and other nano-sized particles
- Research attention was paid to LII starting from the early 90's

History of LII

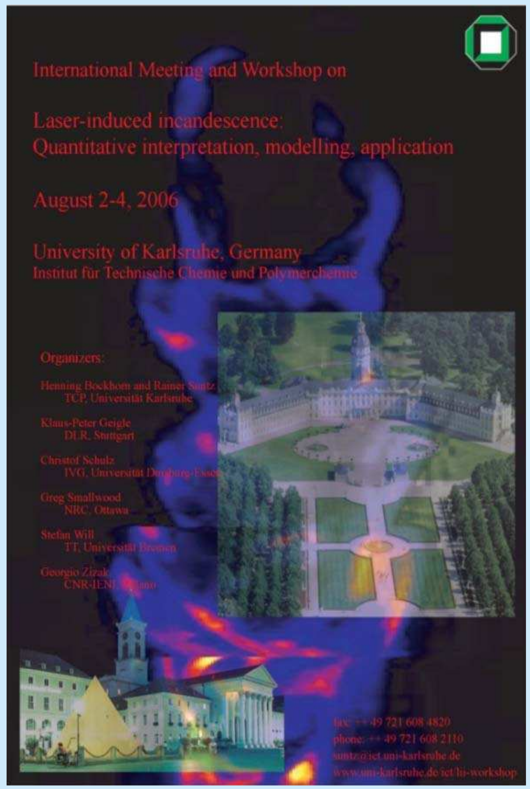
- Early research and applications were in Europe, US, and Canada
- It has gained increased attention in Japan, South Korea, and China
- **The driving force for the popularity of LII is the concerns of soot emissions from various combustion devices and the need to advance our fundamental understanding of soot formation**

History of LII

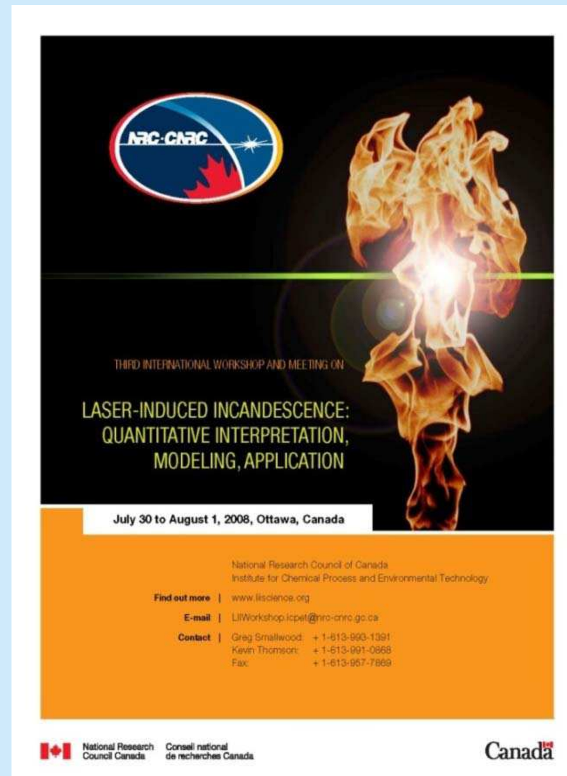
- **Three** international LII Workshops have been held in the last few years (www.liiscience.org)



2005



2006



2008

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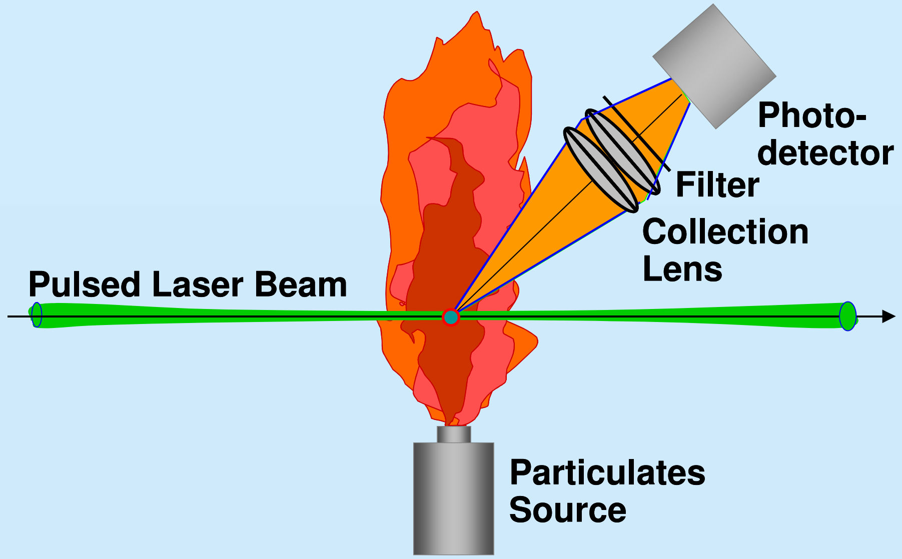
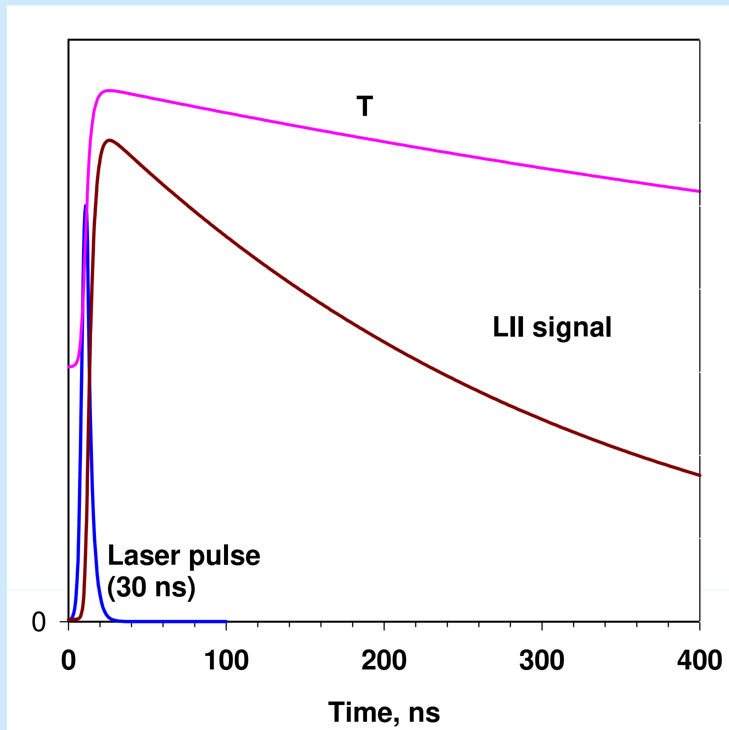
Principle of LII

- LII experiment:
  - pulsed laser beam (ns)
  - rapid heating of soot to about 4000 K
  - soot radiates incandescence while cooling to ambient temperature
  - incandescence signal is collected to determine soot concentration, specific surface area, and primary particle diameter



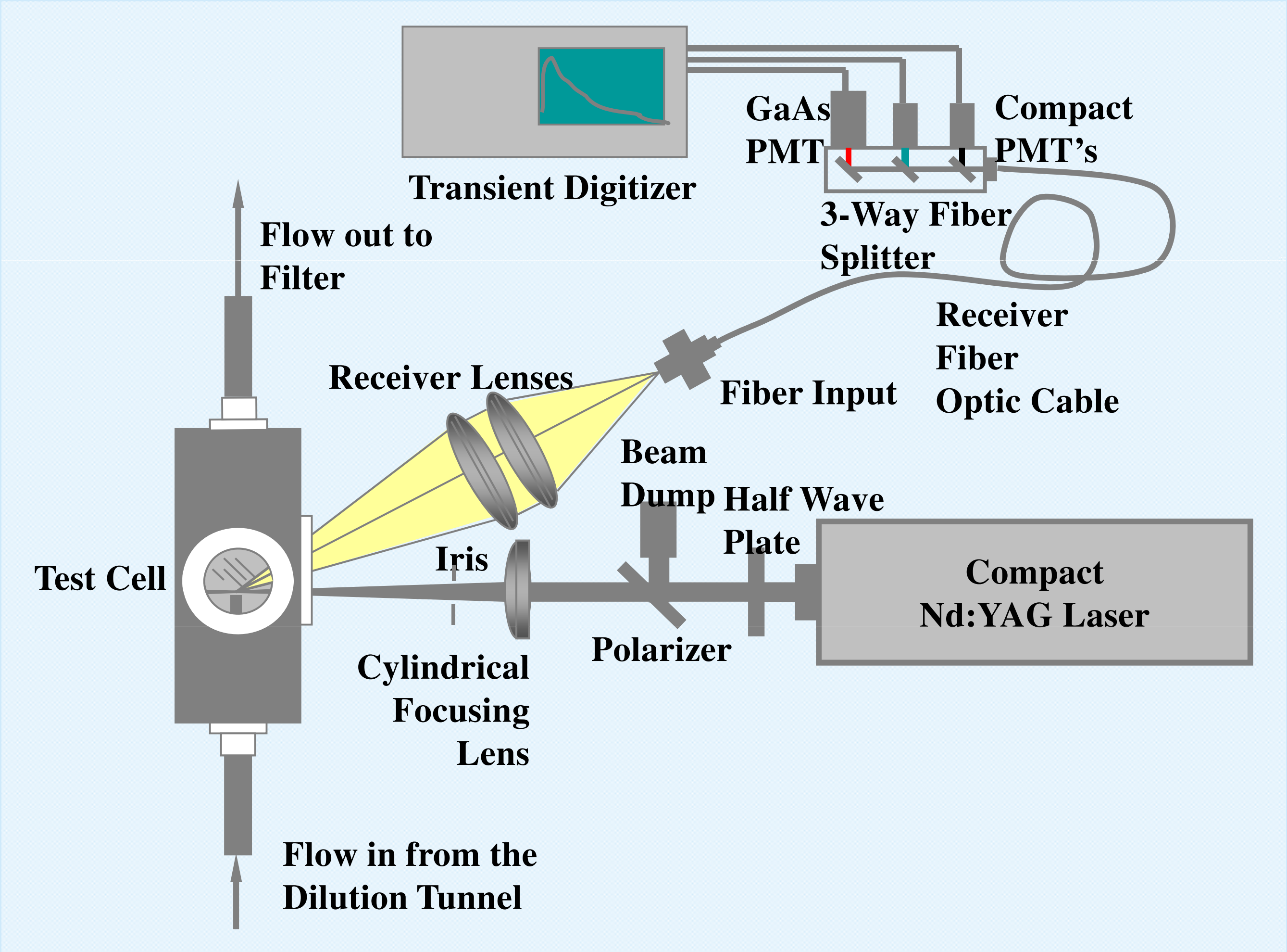


$$f_v \propto \text{LII}_{max}$$

LII Set-ups

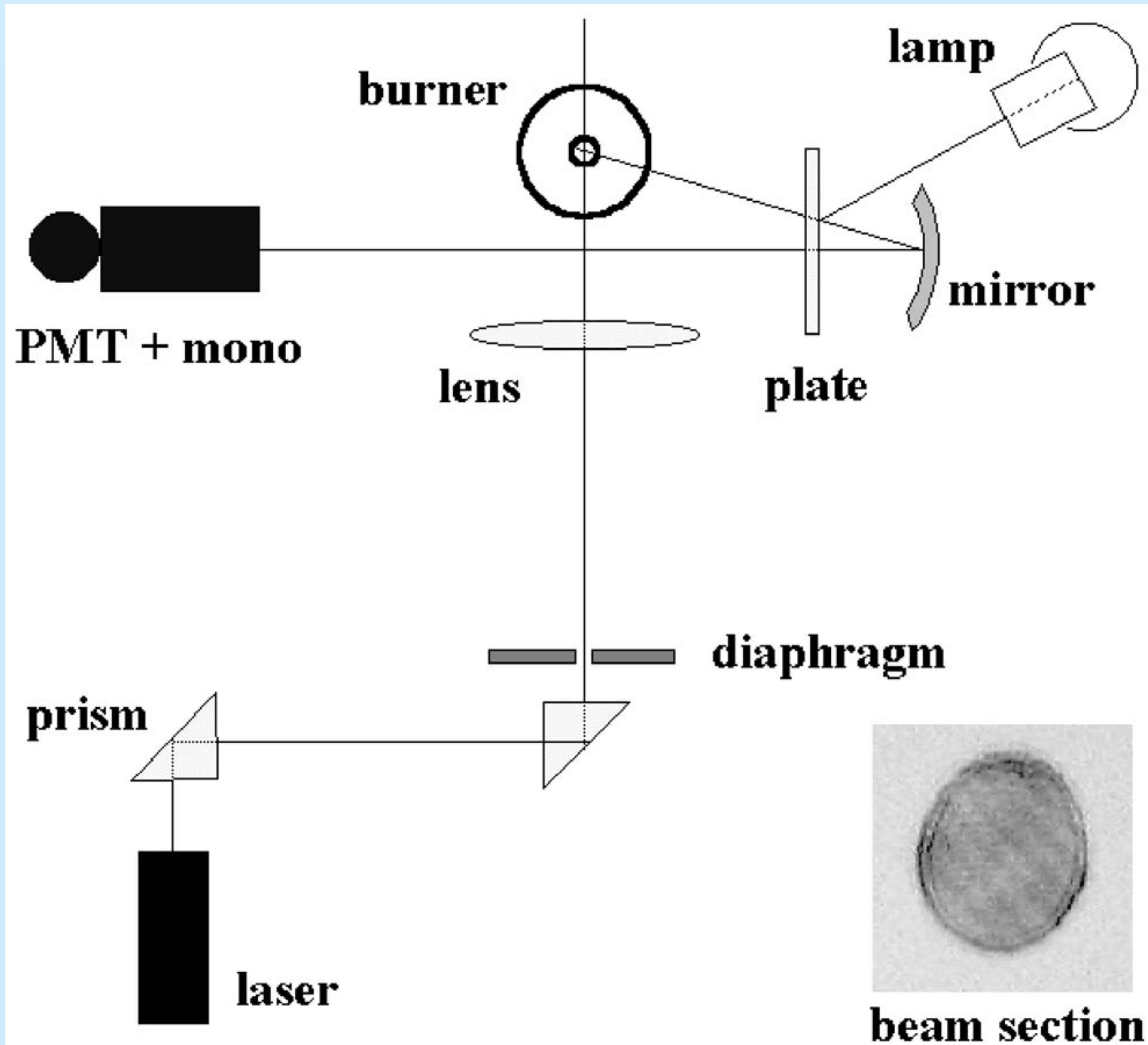
- LII components: **Laser source**, particle source (flame or generator),**signal collection system**

NRC
Set-up

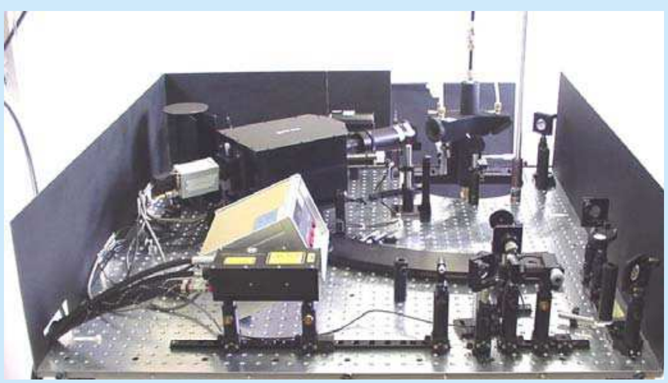


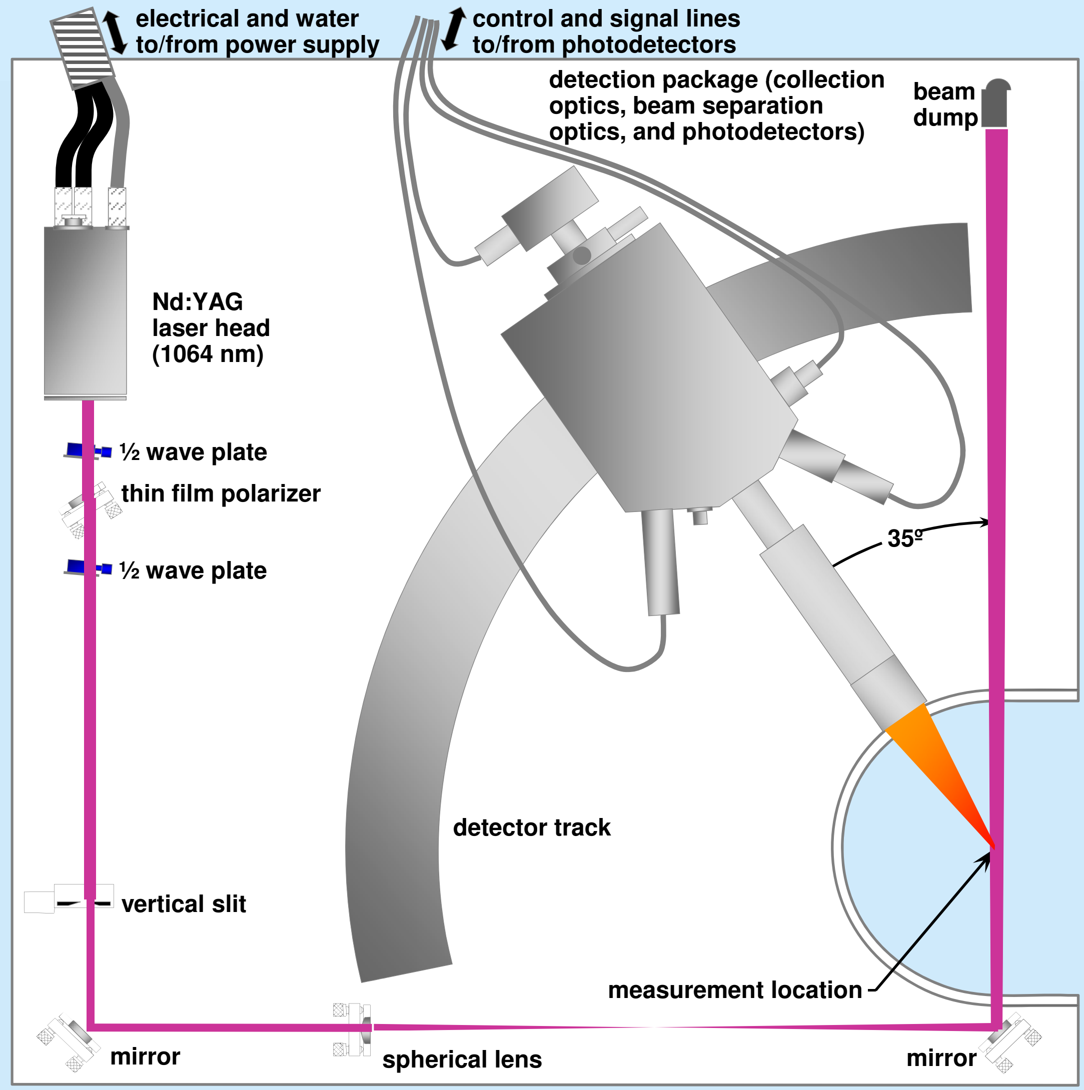
LII Set-ups

CNR (Italy)
Set-up



NRC LII Apparatus

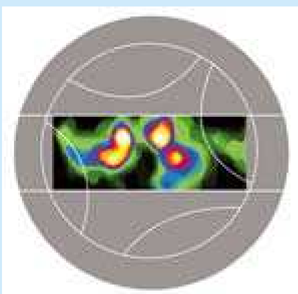




Commercial LII Devices

La Vision

http://www.lavision.de/techniques/lii.php



soot formation in a Common Rail 2l diesel

Applications:

- in-situ and real time characterization of soot emission in diesel and direct injection spark ignition engines, gasturbines, flames and various kinds of metal or ceramics
- particle flows

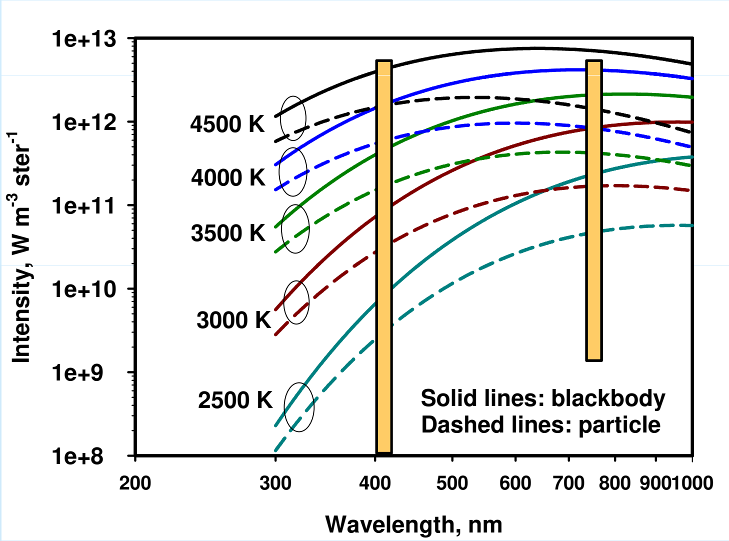
Artium

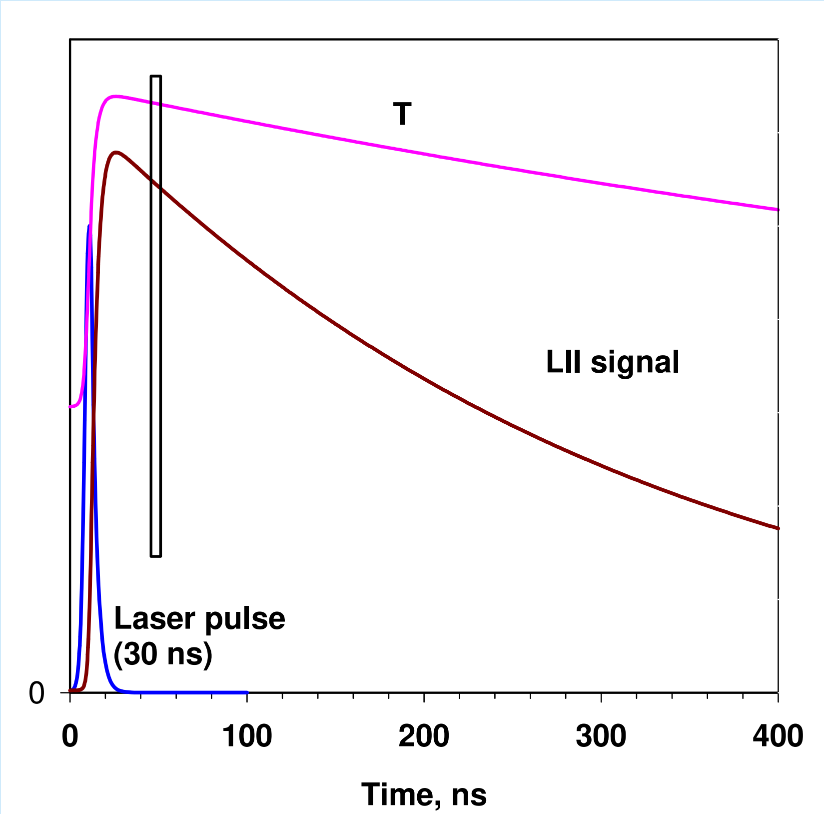
http://www.artium.com

Issues for LII System

- Excitation wavelength (1064 nm or 532 nm)
- Laser fluence choice: **high** or **low**
- Detection issues: wavelength, gated or time-resolved, gated width and timing
- Point LII (PMT) or 2D LII (ICCD)
- Calibration:
  (1) conventional approach
  $f_v = C\ S_{LII}$
  (often through comparison with laser extinction)

  (2) absolute intensity approach
  (detect absolute LII intensity and particle temperature)
  (particle temperature is measured through two-color LII detection)





Features of LII

- Relatively easy to implement
- Non-intrusive (?)
- Very high temporal resolution (turbulent flames, engine combustion)
- Very high dynamic range (ppb up to ppm)
- Good spatial resolution for point measurement
- 2D setup (soot imaging in laminar and turbulent flames, engine cylinder)
- Provides soot volume fraction and primary particle size

- However, it requires sophisticated theory to quantitatively understand the processes to determine primary particle size (distribution)

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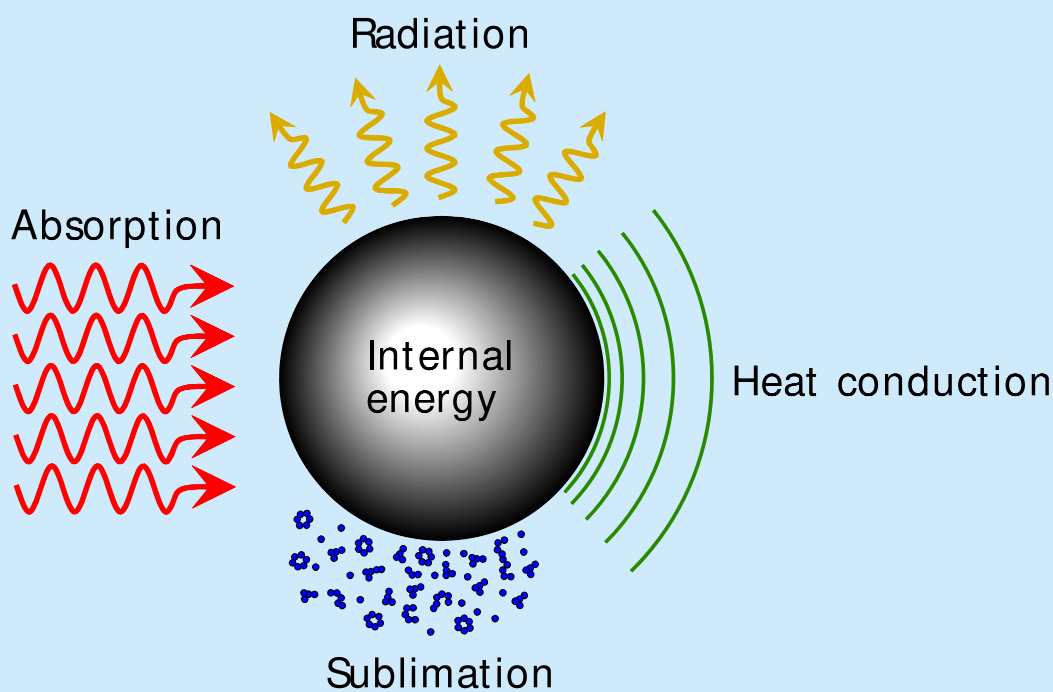
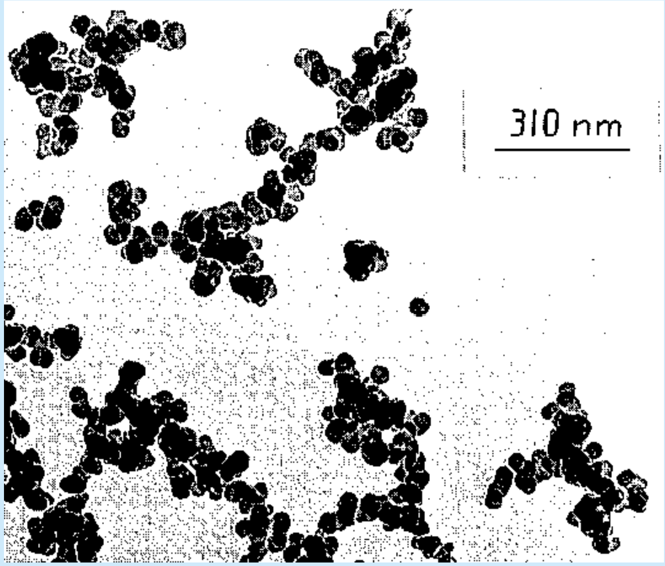
Theory of LII

- LII theory
  - Numerical model of nanoscale (temporal and spatial) heat and mass transfer to and from the particles
  - LII theory is essential to interpret the detected signals, especially for time-resolved LII to obtain particle size information



Structure of soot

- Need to know the structure of soot to establish LII theory
- How soot particles look like?
  - form fractal aggregates
  - not isolated spherical particles
- Primary particles are more or less uniform in size



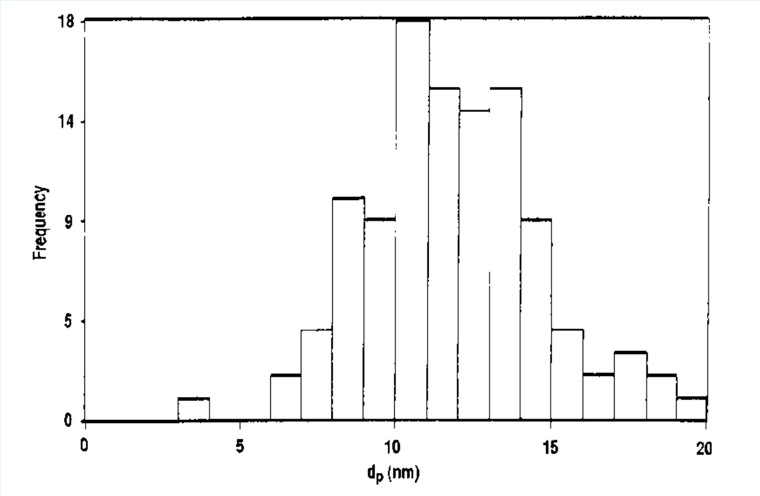
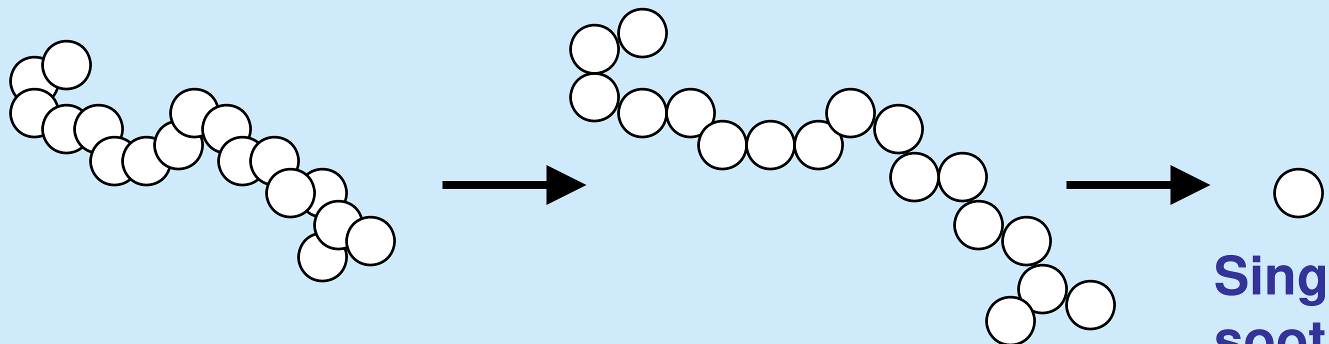


FIGURE 2 Histogram of primary particle diameters of soot aggregates collected from the ethene diffusion flame at a height z = 15 mm, and a radial location r = 3.1 mm.

Megaridis and Dobbins, Combust. Sci. Tech. Vol.71, pp.95-109, 1990

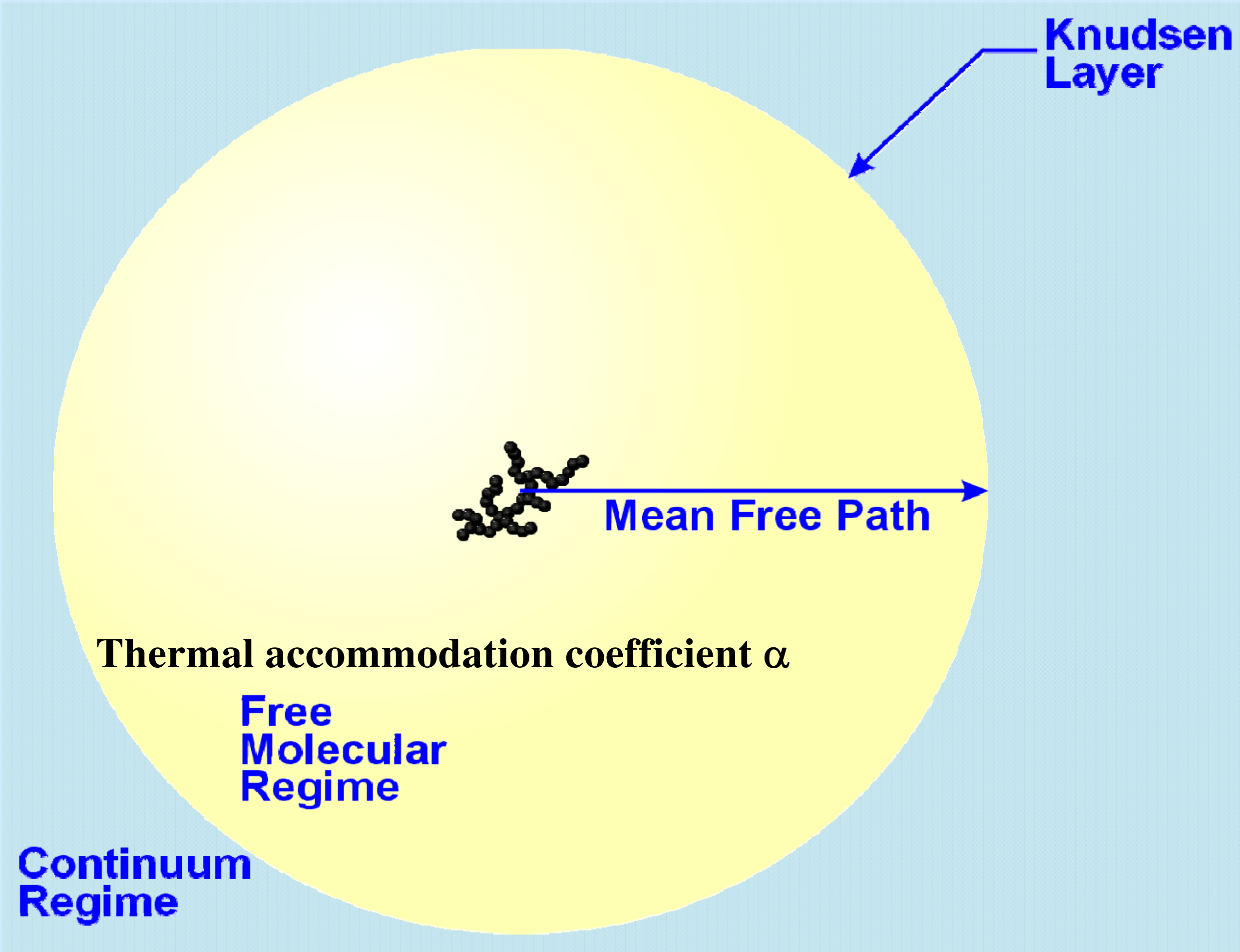
Conventional LII Model: single particle based

- Assumptions: neglect the effect of aggregation on particle laser absorption and particle cooling



LII Theory

- LII model: schematic length scales for soot in flames



1 atm
T = 1700 K
MFP: 600 nm

LII Theory

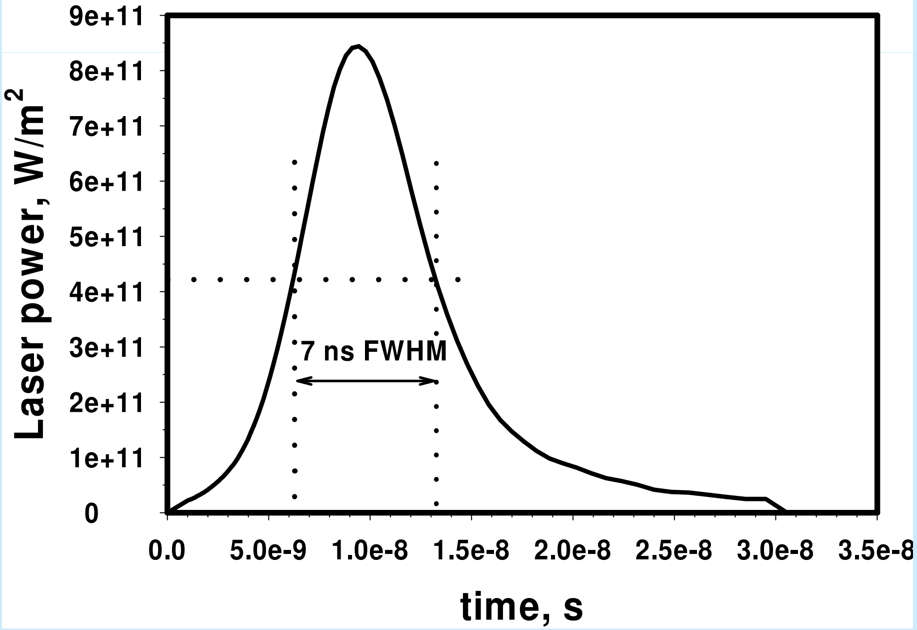
- Time and length scales for laser absorption

Primary soot particle: 30 nm
Laser wavelength: 1064 or
532 nm
Heat diffusion time:

$$\tau_d = \frac{d_p^{\ 2}}{k_s / \rho c_p}$$

which is in sub-nanosecond

⟶ **Particle is isothermal**



$\pi d_p / \lambda$ **is a small value (~0.1), i.e. Rayleigh regime**
Absorption and emission is a volumetric process

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LII Theory

- Energy Equation for a Single Primary Soot Particle

$$\underbrace{\frac{\pi D^3}{6}\rho_s c_s \frac{\mathrm{d}T}{\mathrm{d}t}}_{\mathrm{I}} = \underbrace{C_a q}_{\mathrm{II}} - \underbrace{\frac{2\ k_a \left(T - T_0\right)\pi D^2}{\left(D + G\ \lambda_{MFP}\right)}}_{\mathrm{III}} + \underbrace{\frac{\Delta H_v}{M_v}\frac{\mathrm{d}M}{\mathrm{d}t}}_{\mathrm{IV}} + \underbrace{q_{rad}}_{\mathrm{V}}$$

I	particle internal energy variation rate
II	laser heating
III	heat transfer to surrounding gas
IV	soot evaporation
V	radiative heat loss

$$C_a = \frac{\pi^2 D^3 E_m}{\lambda}$$

$$G = \frac{8f}{\alpha(\gamma+1)}$$

$$f = (9\gamma - 5)/4$$

Large uncertainty in E_m and α exists

- Other physical processes neglected, such as photodesorption, annealing, and oxidation

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LII Theory

- Mass Transfer Equation for Soot Particle

$$\underbrace{\frac{\mathrm{d}M}{\mathrm{d}t}}_{\textbf{I}} = \frac{1}{2}\rho_s \pi D^2 \frac{dD}{dt} = \underbrace{-\pi D^2 N_v \frac{M_v}{N_A}}_{\textbf{II}}$$

I	particle mass loss rate
II	vaporization rate

$$N_v = \beta \frac{P_v N_A}{RT} \sqrt{\frac{RT}{2\pi M_v}}$$ molecular flux

LII Theory

- Radiation loss term

$$q_{rad} = 8\pi^3 D^3 E(m) \frac{k^5}{h^4 c^3} T^5 \int_0^\infty \frac{t^4}{e^t - 1} dt$$

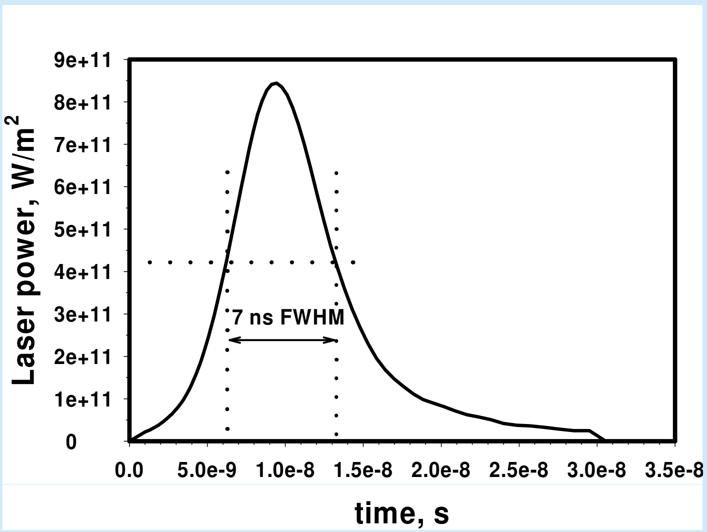
- LII signal intensity

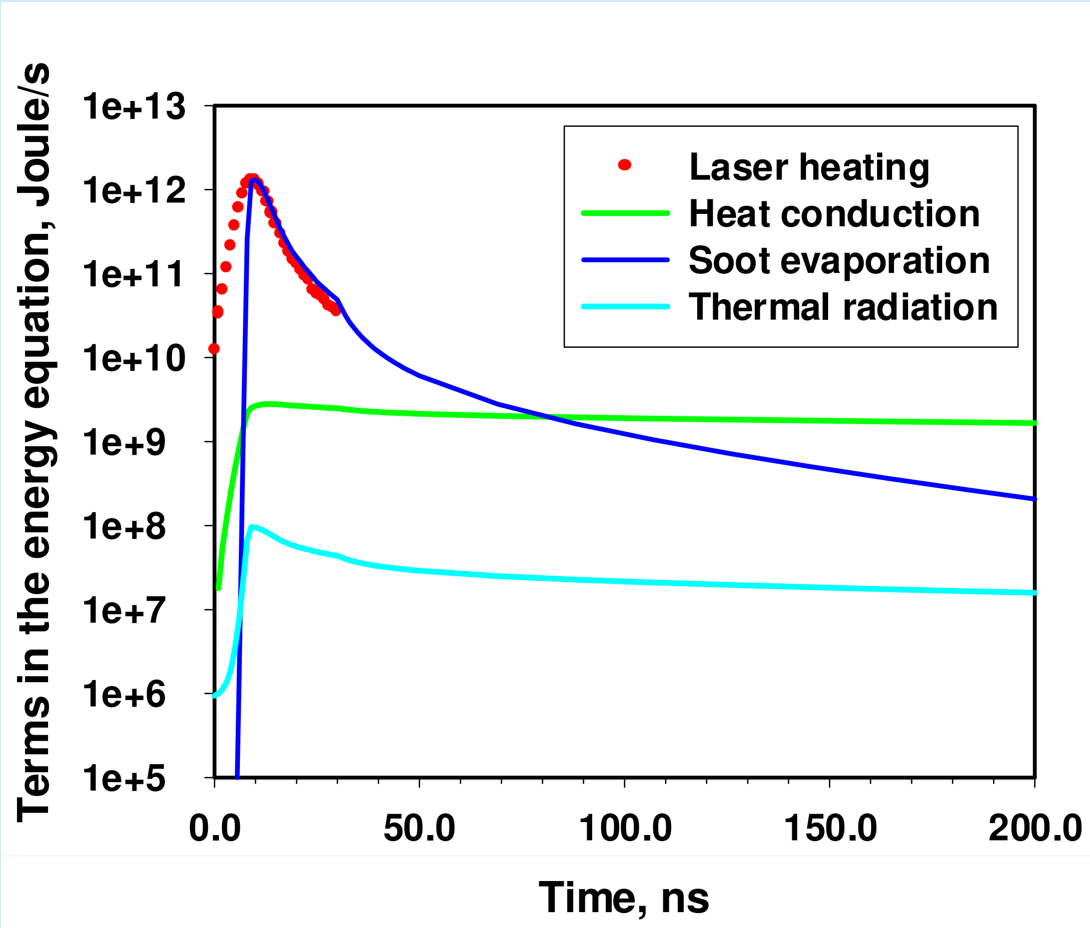
$$S_{LII} \propto \frac{8\pi c^2 h E(m)}{\lambda^6} D^3 \exp(-\frac{hc}{\lambda k T})$$

Some numerical results

- Relative importance of heating and cooling terms
  Tophat spatial profile at 6 mJ, d_p = 32 nm, E(m) = 0.261.

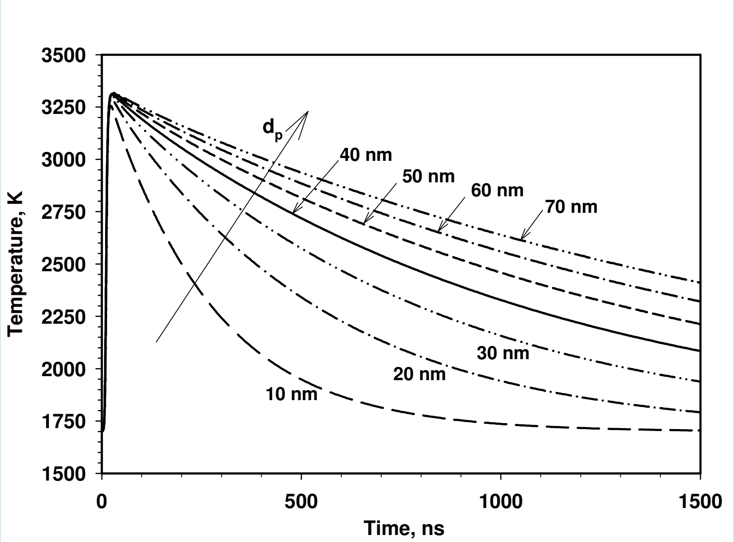
532 nm

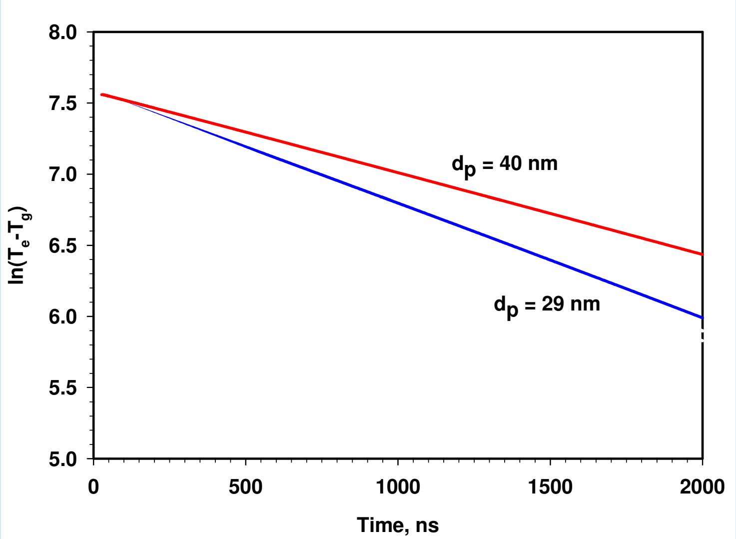




Particle sizing strategies

- Larger particles cool slower than smaller ones:
  Internal energy $\propto d_p^3$
  Conduction cooling $\propto d_p^2$
- The time-resolved LII signal carries information about the particle size
- Particle temperature can be inferred from two-color or multi-color LII detection
- The Temp decay rate is inversely proportional to d_p





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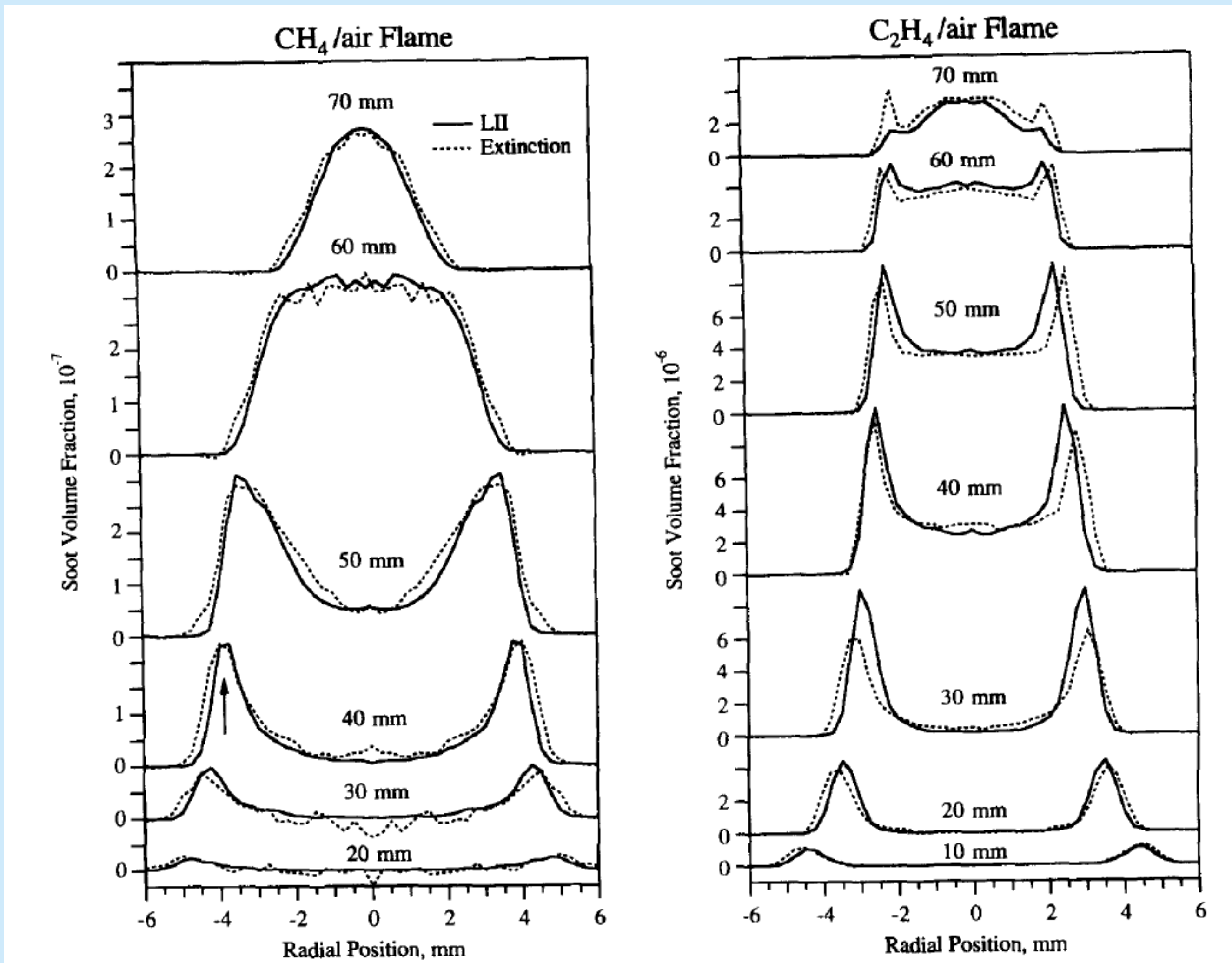
Capabilities of LII

- Steady and flickering laminar diffusion flames



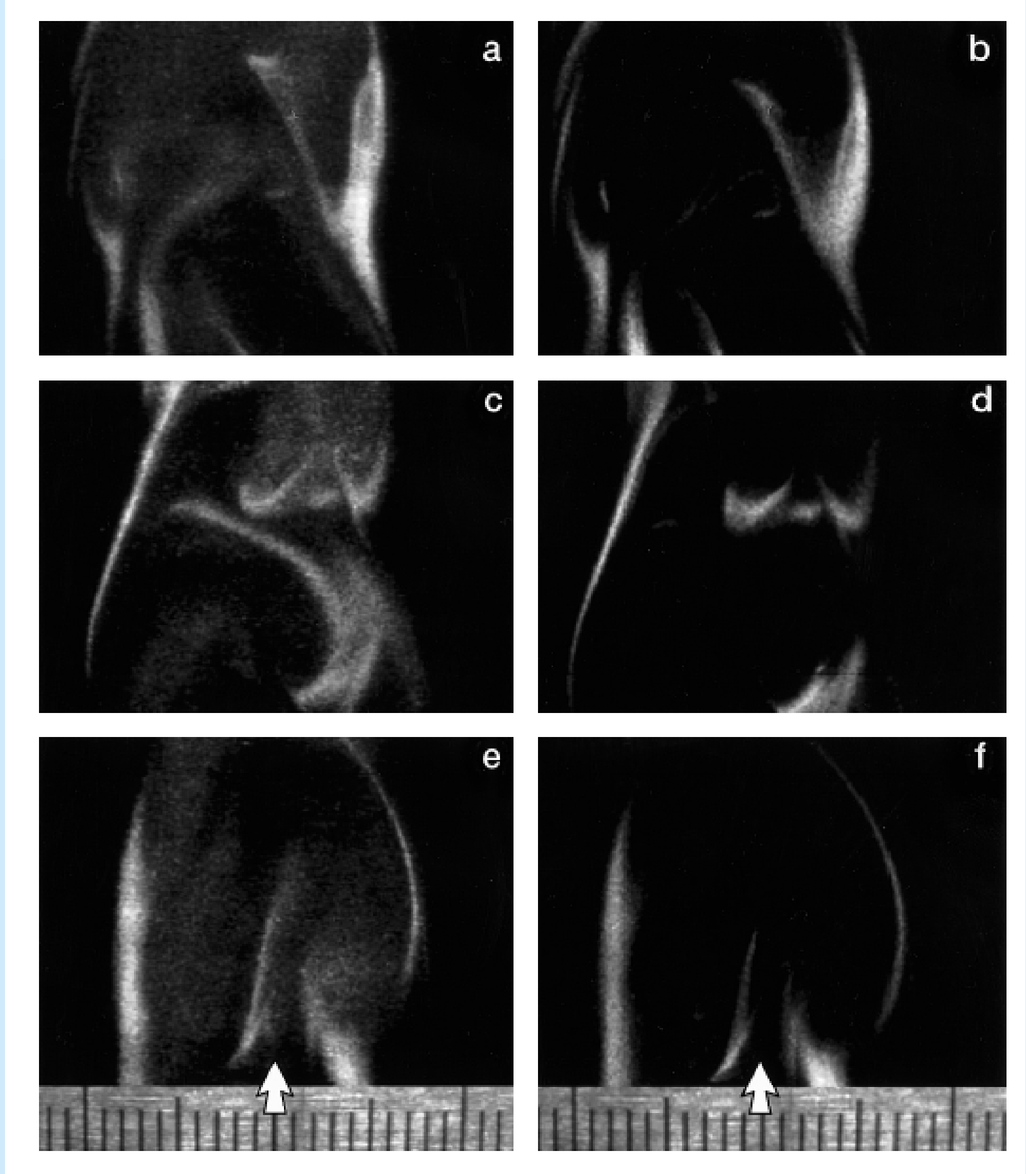
Shaddix and Smyth, CNF 1996

Capabilities of LII

- Turbulent jet diffusion flames (ethylene)

7.6 to 9.5 cm
above the burner

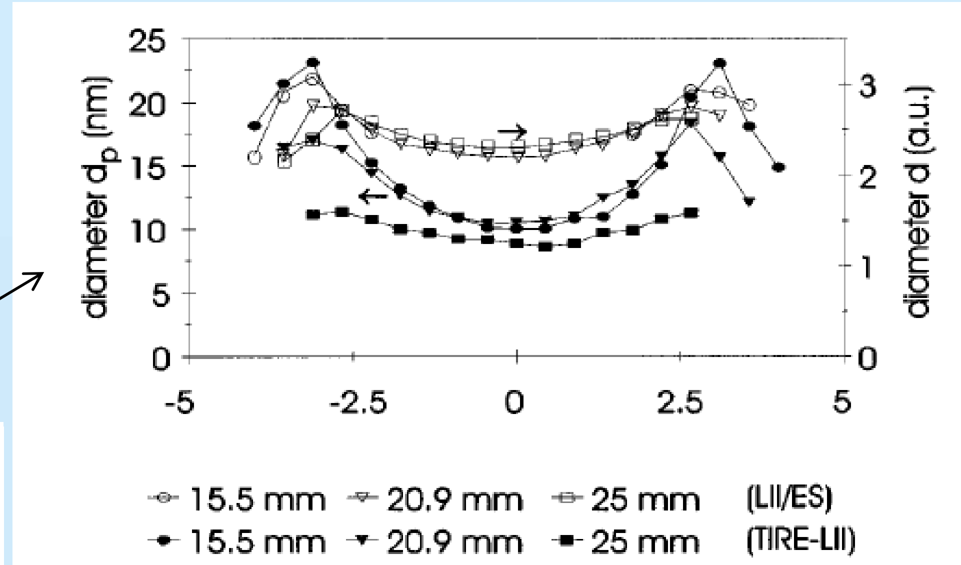
266 nm 1064 nm



Vander Wal, Exp. In Fluids 1997

Capabilities of LII

- Laminar diffusion flames (d_p)



Will et al., Optical Letters, 22, pp. 2342-2344, 1995.

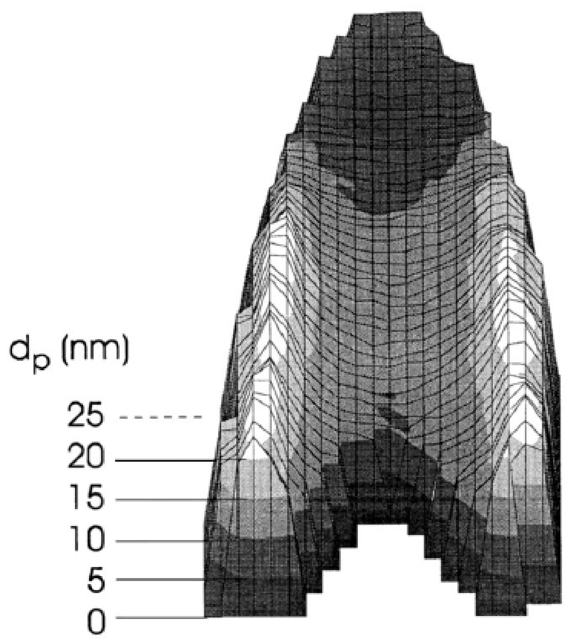
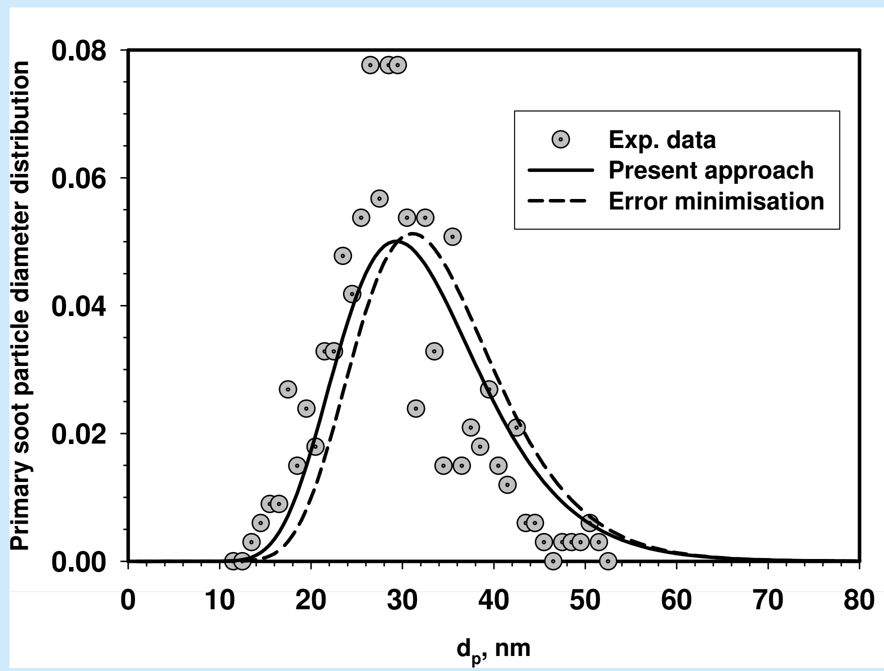


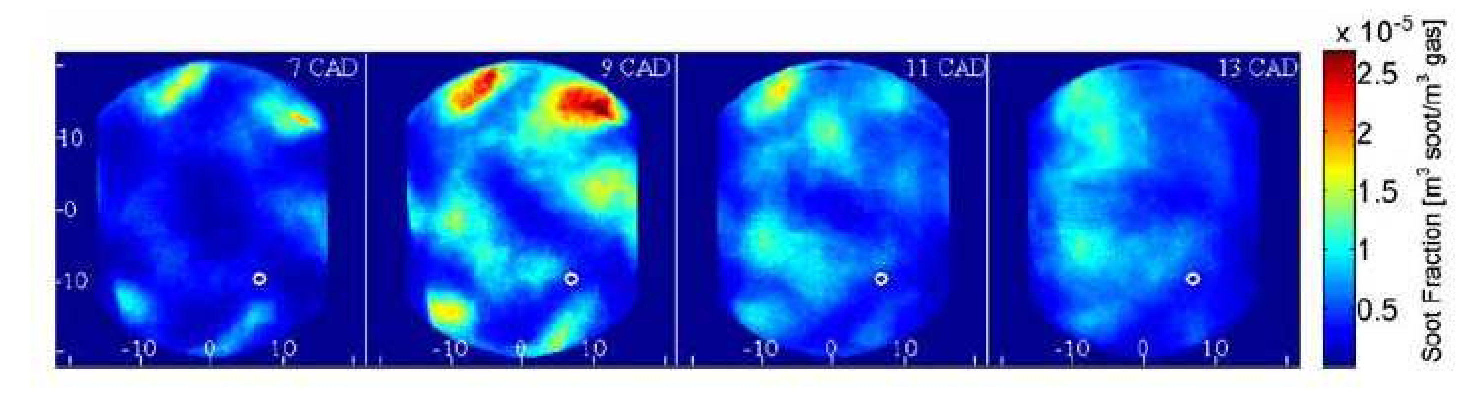
Fig. 3. Three-dimensional representation of the distribution of primary particle sizes within the observation plane. The image was acquired in a laminar ethene–air diffusion flame by time-resolved LII.



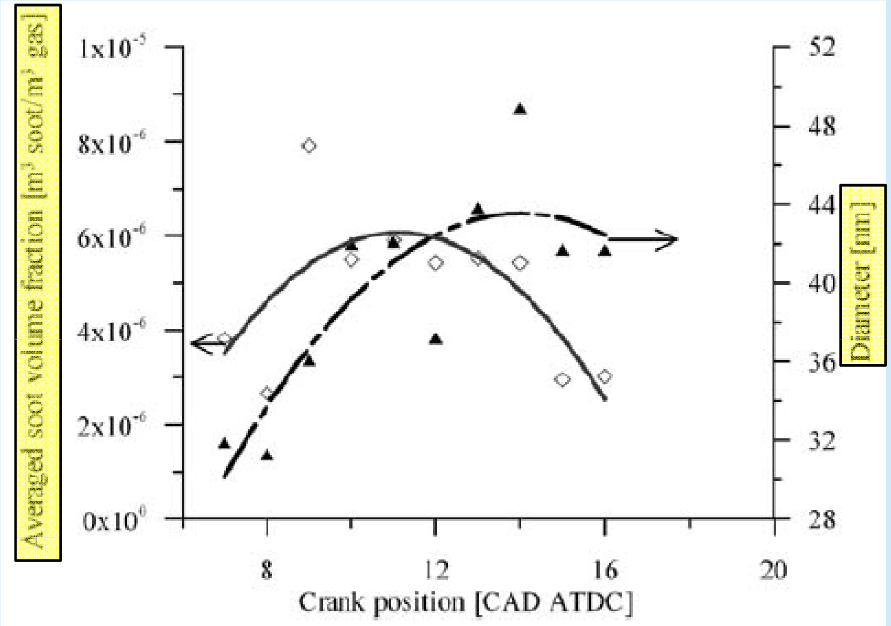
Liu et al., Int. J. Heat Mass Transfer, 2006

Capabilities of LII

- 2D Two-Color Time-Resolved LII (f_v and d_p)



Averaged 2D soot fraction distribution inside an engine combustion chamber. From left to right: 7,9,11,13 CAD ATDC



Mean soot volume fraction and local mean diameter evolution for different crank angle

Boiarciuc et al., Appl. Phys. B, 83: 413-421 (2006)

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Summary of LII Applications

- LII has become the preferred method for soot measurement in many areas of combustion:
  - laminar flames (atmospheric and elevated pressures)
  - shock tube
  - droplet combustion
  - in-cylinder engine combustion
  - turbulent flames
  - engine exhaust
  - particulate concentration in environment (high sensitivity LII)

- LII has been applied to carbon black industry to do real-time monitor of particle size and to medical applications

- LII has also been used to other non-carbon refractory particles, such as metal oxides, synthesized in flames

Opportunities for LII in Soot Studies

- Capabilities of LII: soot volume fraction, primary particle size, primary particle number density, the latter is unique to LII and is critical to understanding soot formation

- LII provides very high temporal resolution and very wide dynamic range

- LII can be combined with LIF and Laser Scattering to obtain further information on soot formation processes, such as relative spatial locations of PAHs and soot and soot aggregation

- **These capabilities make LII a powerful tool to advance fundamental understanding of soot formation and to investigate soot in turbulent flames and in engine combustion**

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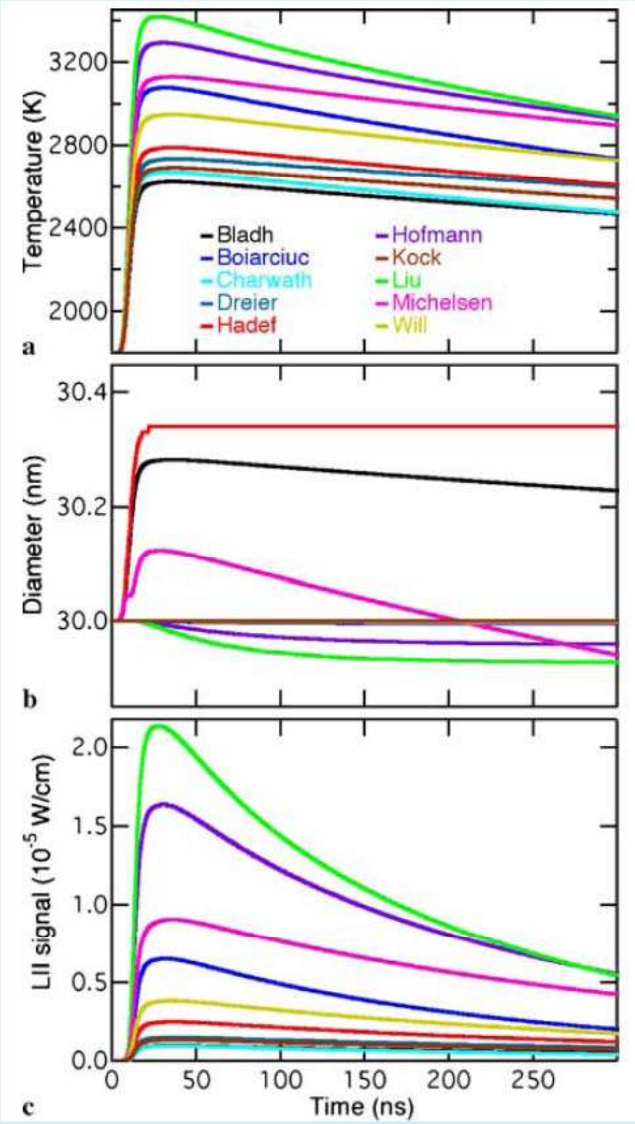
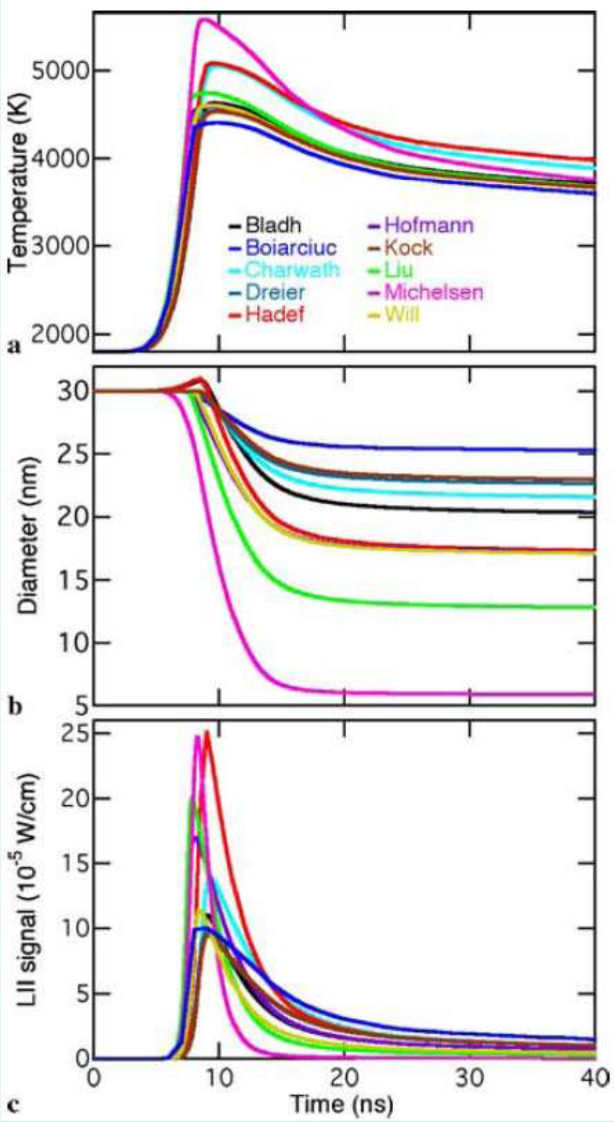
How different LII models compare?

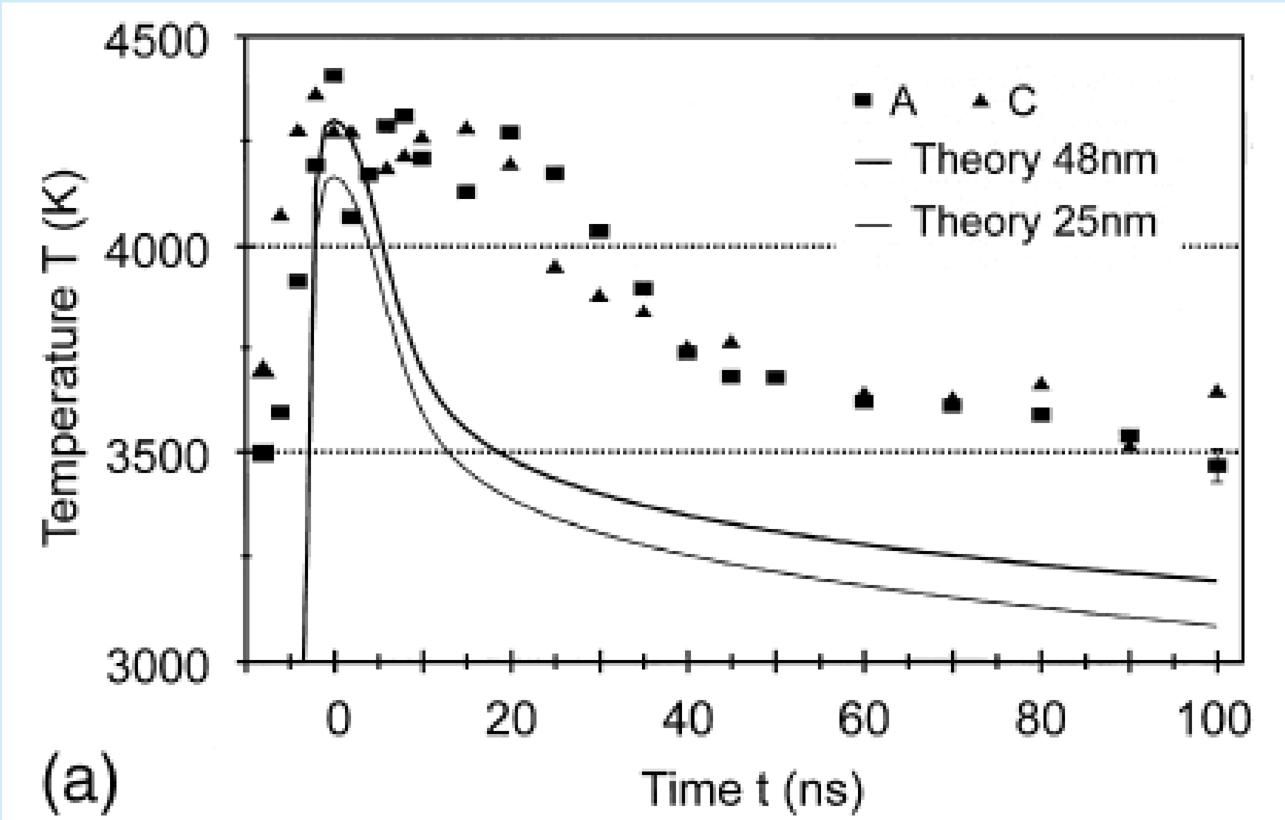
- Comparison of different LII models (Michelsen et al., Appl. Phys. B, 2007)





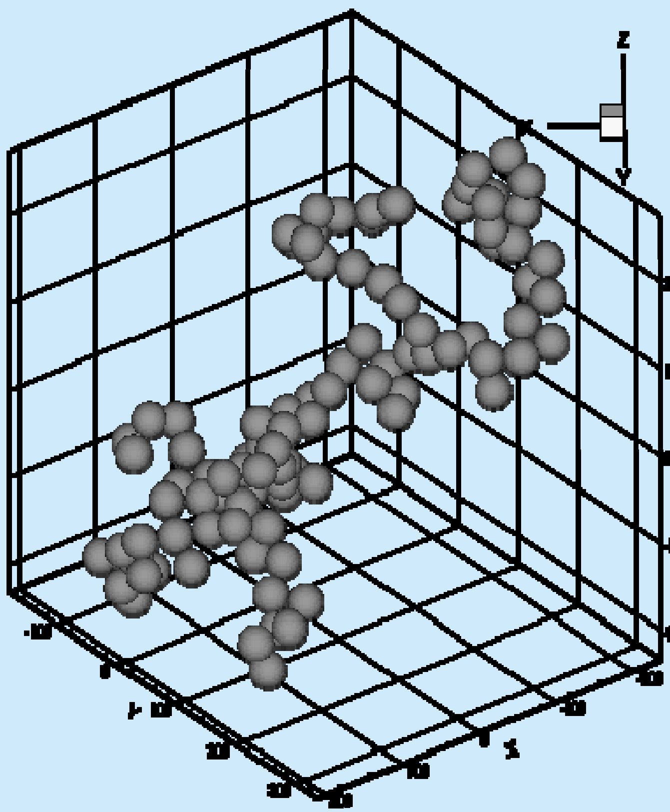
How does LII model perform in high-fluence?

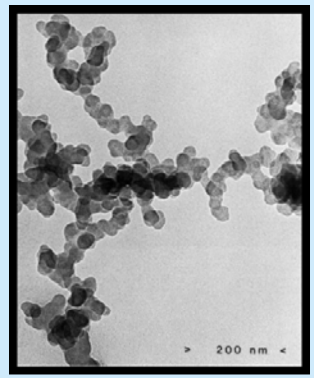
- Schraml et al., Combustion and Flame 2000

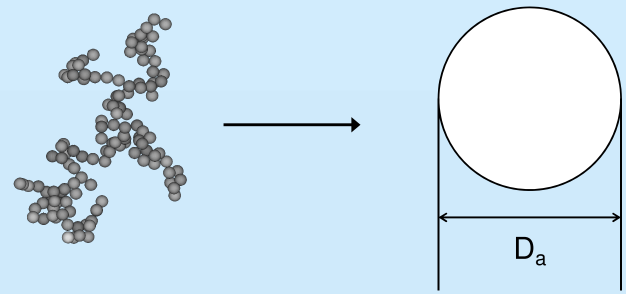


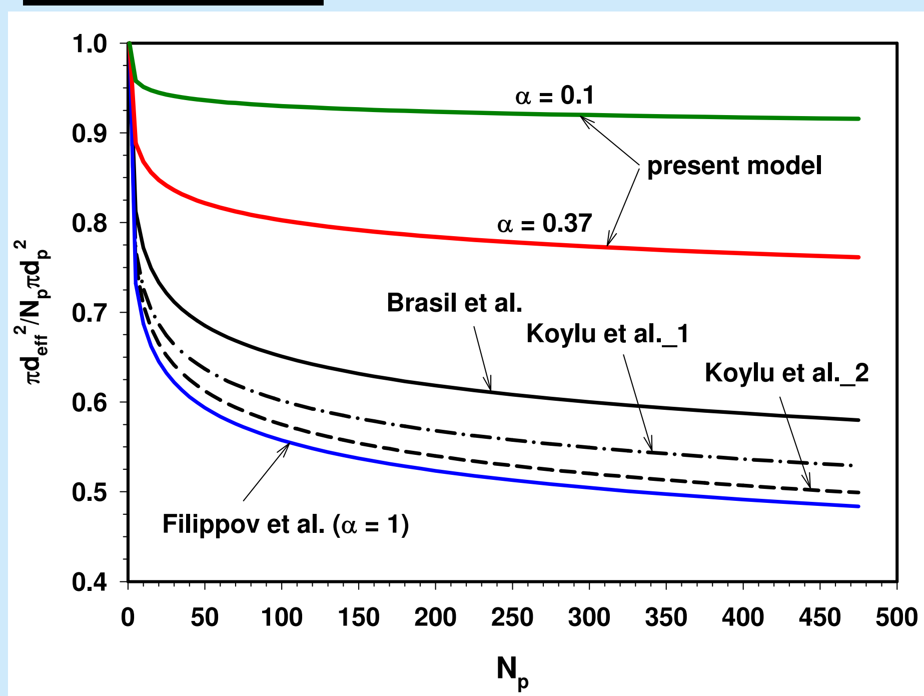
Need to consider the fractal structure of soot

- Three-dimensional display of a soot aggregate (129)





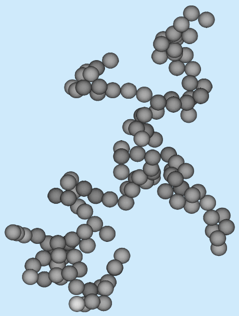


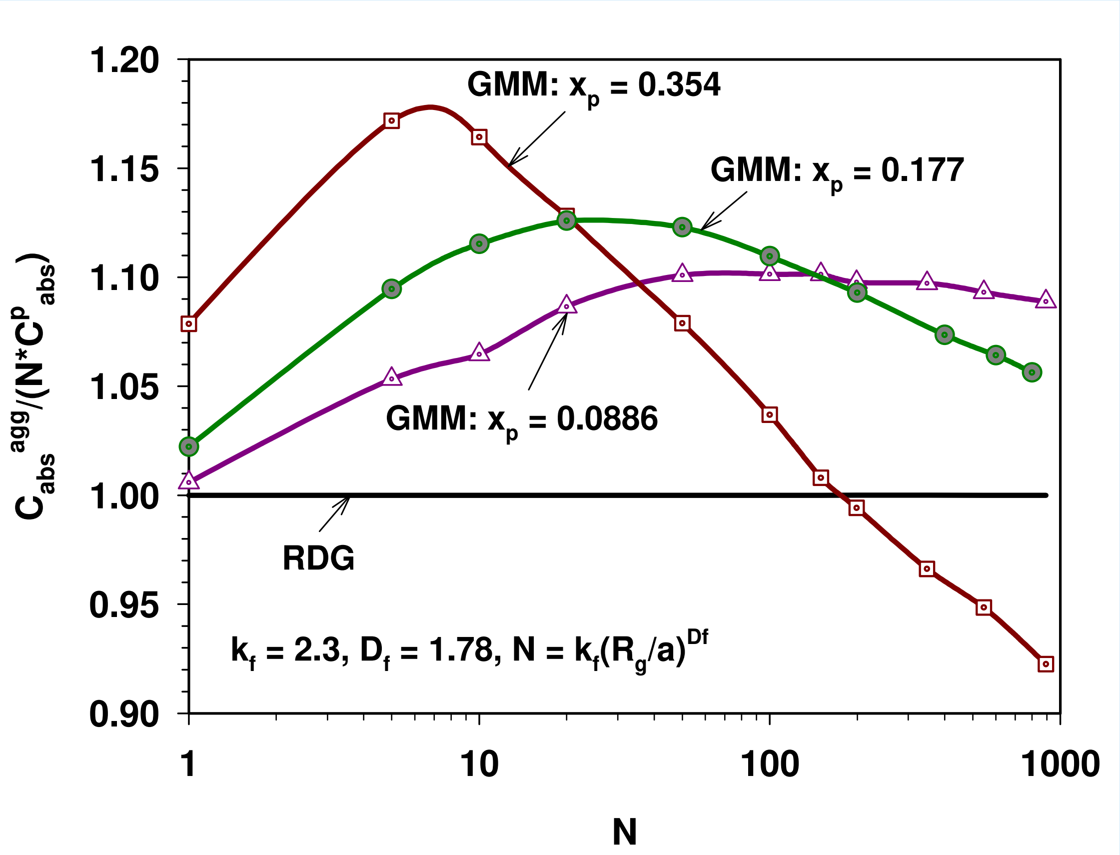


DSMC is also capable of calculating the sublimation

Need to consider the fractal structure of soot

- Is Rayleigh-Debye–Gans approximation acceptable for LII application?





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Remaining Challenges

- How important is C_2 and other non-carbon emissions to LII signal detection when 532 nm laser is used?
- Improvement over 2D two-color time-resolved LII for volume fraction and particle size measurements
- Shot noise
- Selection of laser fluence, detection wavelength(s)
- Is LII signal truly proportional to soot volume fraction?

$$S_{LII} \propto d_p^{3+154nm/\lambda}$$

- Well controlled LII experiments for model validation

Remaining Challenges

- Uncertainty in soot refractive index and thermal properties at ~ 4000 K
- How important are other physical and chemical processes? (annealing, photodesorption, soot oxidation)
- How accurate is the soot sublimation model for a single particle?
- How to account for the effect of particle aggregation on soot sublimation?

Outline

- Background
- A brief history of LII
- Introduction of LII: principle, setup, issues
- LII Theory
- LII applications in combustion: capabilities and opportunities
- Remaining challenges
- Conclusions

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Conclusions

- LII offers many advantages over conventional techniques and has evolved into a powerful diagnostic tool for soot measurement in combustion applications
- Its ability to infer primary soot particle size could potentially make significant contributions to advance our fundamental understanding of soot formation
- The current single particle based LII model is reasonable for inferring primary particle size in the low-fluence regime and under flame conditions
- Soot sublimation is still a poorly understood process and hinders our quantitative understanding of LII in high-laser fluence regime

Conclusions

- Particle aggregation should be accounted for in LII theory
- LII alone cannot provide sufficient information to determine soot morphology; Combined LII/LS is a promising technique to obtain more complete information about morphology
- Many unanswered questions remain in the LII theory and practice to make LII more accurate and more reliable, which offer a great research opportunity

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