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Istituto di Chimica e della Materia
Condensata e di Tecnologie per l'Energia

A look at soot optical properties during laser-induced incandescence measurements

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7th ISF Workshop
on the Measurement and Computation
of Reacting flows with Carbon Nanoparticles

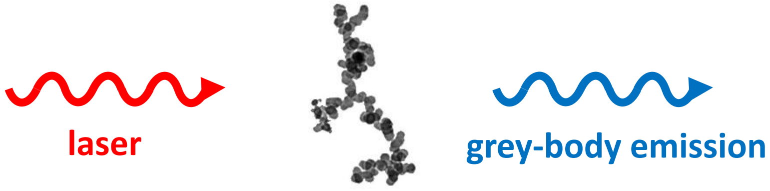
July 20th-21st, 2024 - Milan

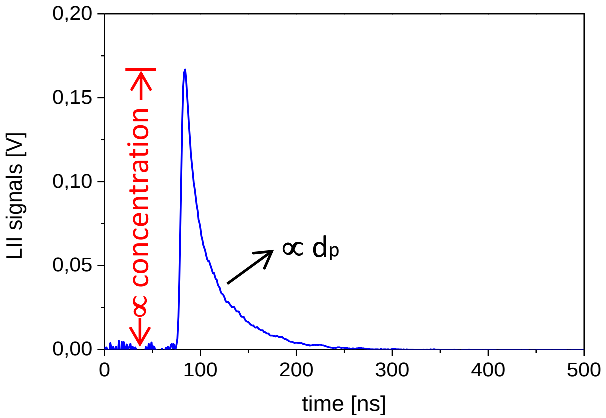
Outline

* Brief recall of laser-induced incandescence (LII) technique
* Quick overview of recent work on soot optical properties
* Effect of laser irradiation on soot optical properties during LII
* Final remarks and next challenges

Laser-induced incandescence (LII)

LII is based on the irradiation of particles with a high-power pulsed laser and the detection of the subsequent incandescence emission signal





→ Determination of **soot volume fraction**, f_v

Single color/two colors detection

→ Measurements of **primary particle size**

TIRE-LII technique

- ✳ High selectivity for measuring carbon nanoparticles
- ✳ High temporal and spatial resolution → on-line measurements
- ✳ High sensitivity and wide dynamic range → ng/m^3 – g/m^3

Laser-induced incandescence (LII)

Broad spectra of applications:

- ✓ Flames
- ✓ Engines
- ✓ Exhaust gases
- ✓ Ambient atmosphere
- ✓ Engineered nanoparticles
- ✓ Suspensions

Trend in the past two years…

→ employment of 2D configuration (especially in practical systems)
→ application of LII to retrieve E(m)
→ application of LII to study "emerging fuels" (blends/additive/surrogates…)

New challenges

- ✓ LII sensitivity to small/nascent particles
- ✓ LII sensitivity to low concentration in engines/combustors

Main drawback of LII
sensitivity toward the optical properties of the particles

Double dare

1. knowledge of the optical properties of particles with different nature
2. effect of laser irradiation on the optical properties

Optical properties of soot

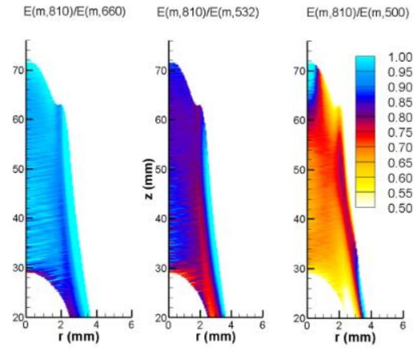
Optical properties can be expressed as:

- ✓ Complex refractive index, **m= n-ik**
- ✓ Soot refractive index absorption function, **E(m)**
- ✓ Dispersion exponent, ***ξ*** or **a**
- ✓ Optical band gap, **Eg**
- ✓ Mass absorption cross section, **MAC**

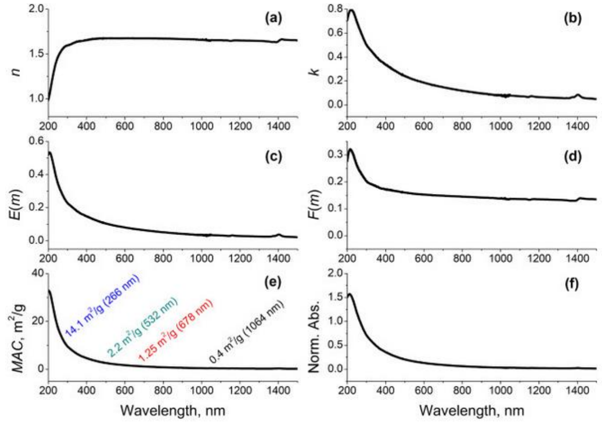
Dependent on:

- ✓ chemical composition/ maturity degree
- ✓ inner structure
- ✓ particle size
- ✓ …

Hot topic in literature

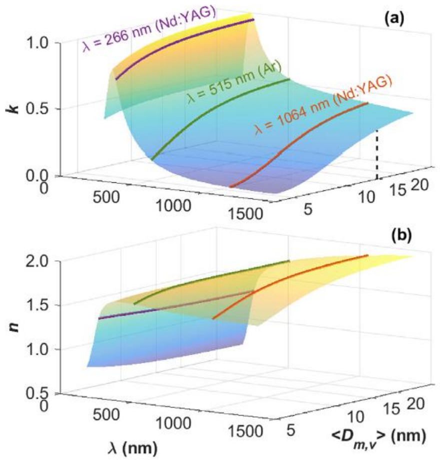


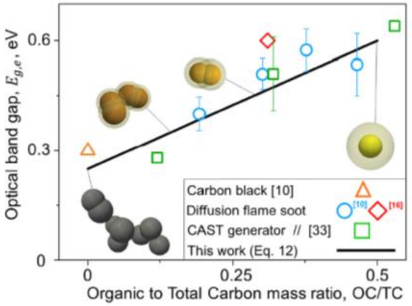
Yon et al.,Combust. Flame 227 (2021) 147–161



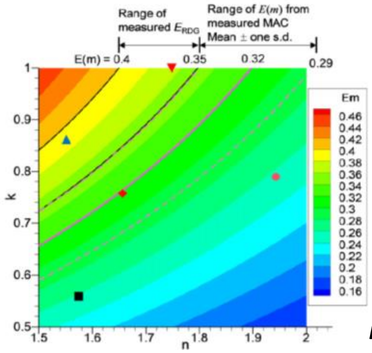
Minutolo et al., Proc. Combust. Inst. 39 (2023) 1129-1138

Wan et al., Proc. Combust. Inst. 38 (2021) 1517-1524





Kelesidis et al.,Carbon 172 (2021) 742-749



Liu et al., Aerosol Sci. Tech. 54 (2020) 33-51

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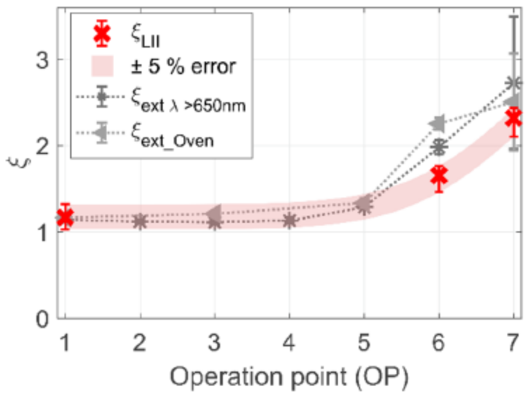
Optical properties from LII

2λ-2C-LII

→ Absorption efficiency of differently matured soot
→ Dispersion coefficient from LII



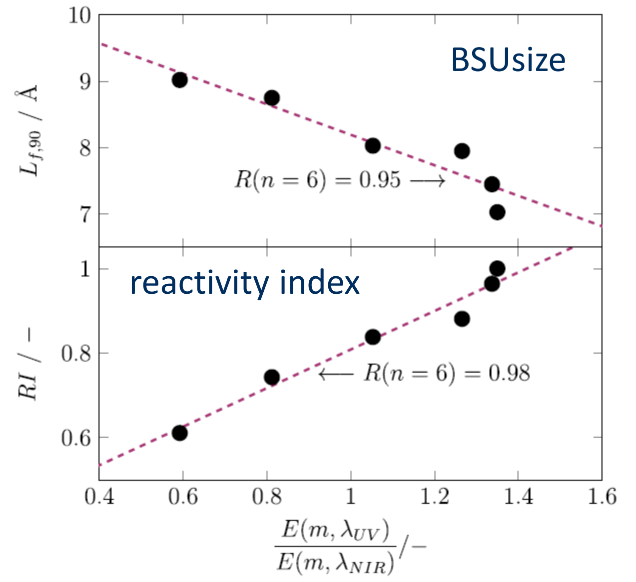
wide spread of *E*(*m, λ*) for soot of different maturity:
E(m, 1064)young=0.05 → E(m, 1064)mature 0.33

Török et al., Appl. Phys. B 127 (2021) 96

Dual-pulse 2C Tire-LII

→ rapid, non-intrusive determination of E(m,λUV)/E(m, λNIR)
→ absolute E(m, λ)

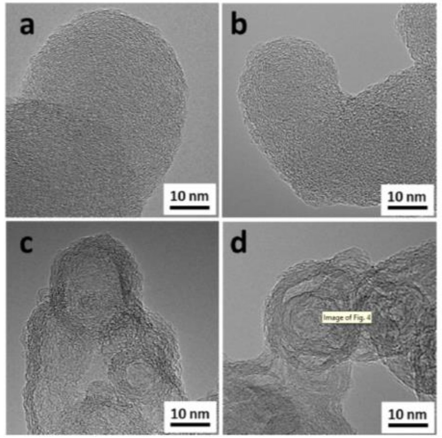
GDI soot



E(m, λUV)/E(m, λNIR) can be used to estimate any nanostructure-associated particle property

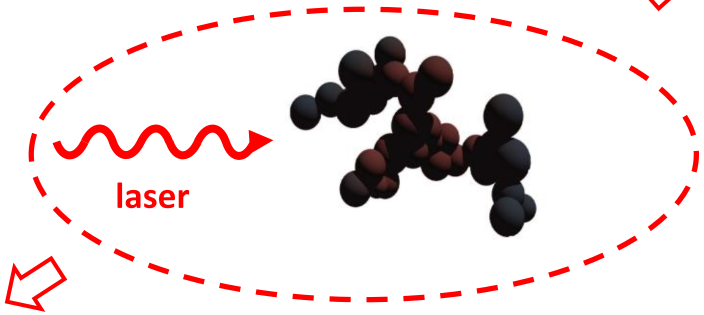
Hagen et al., Combust. Flame 254 (2023) 112850

Effects of laser irradiation on soot particles

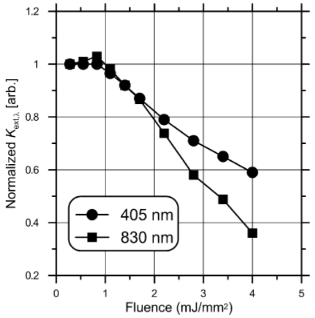


Apicella et al., Combust. Flame 204 (2019) 13–22

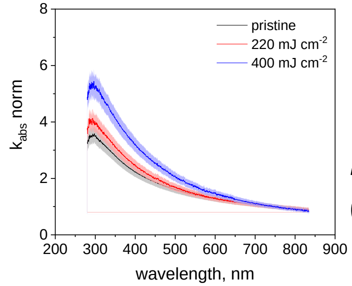
Changes in the particles internal structure



Thomson et al., Appl Phys B 104 (2011) 307-319

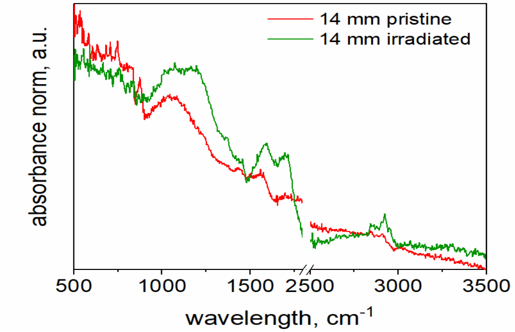


Reversible or permanent change in the optical properties



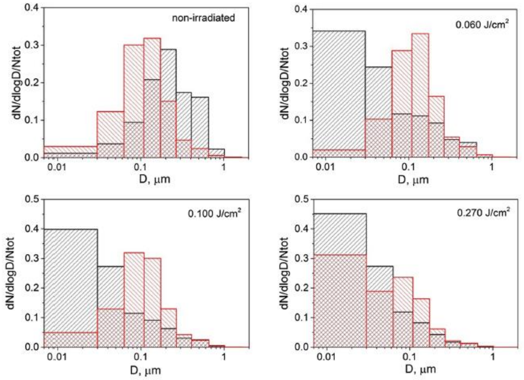
Migliorini et al., Appl Phys B 129 (2023) 129-133

Changes in particles chemistry

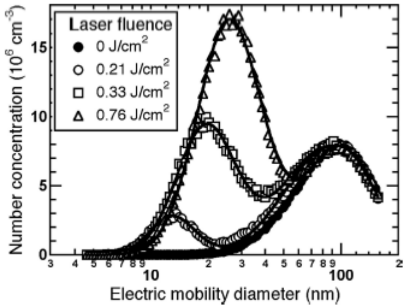


Migliorini et al., under revision

Migliorini et al., Exp. Therm Fluid Sci. 114 (2020) 110064

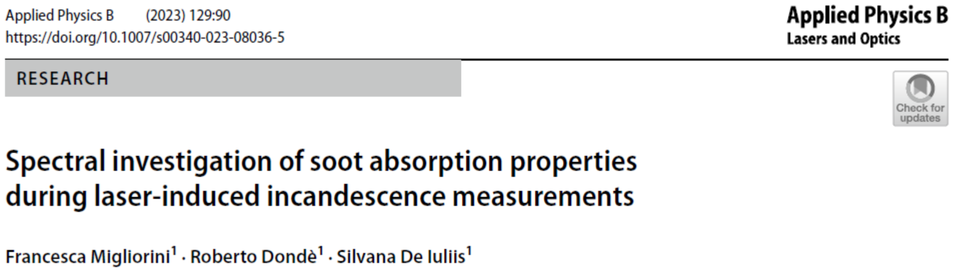


Formation of new particles



Michelsen et al., Appl. Opt 46 (2007) 959-977

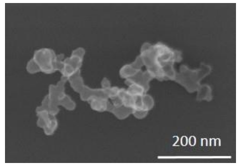
What happens during LII time-scale?


Applied Physics B (2023) 129:90
https://doi.org/10.1007/s00340-023-08036-5
Applied Physics B
Lasers and Optics
RESEARCH

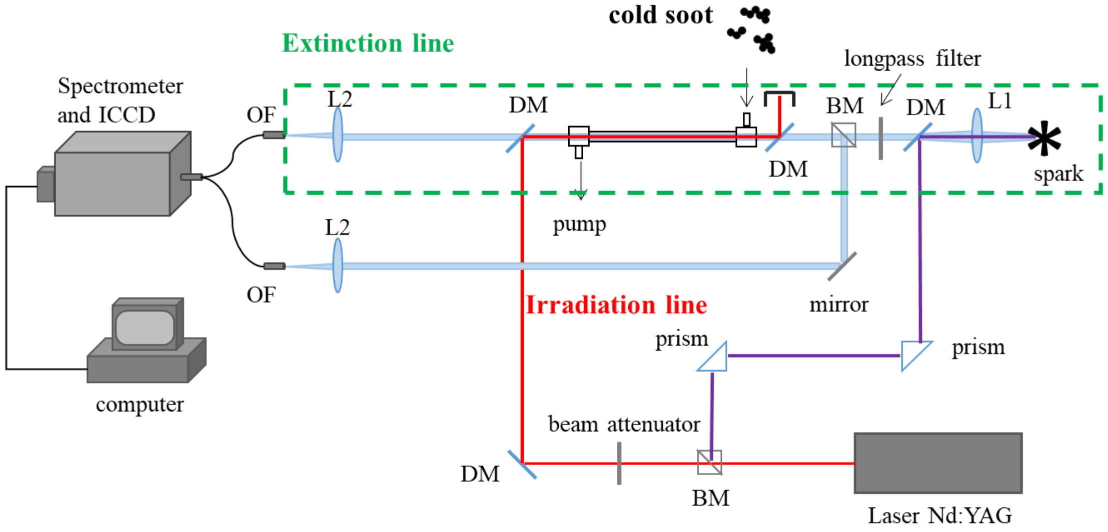
Spectral investigation of soot absorption properties during laser-induced incandescence measurements

Francesca Migliorini[1] · Roberto Dondè[1] · Silvana De Iuliis[1]

Wavelength-resolved extinction measurement during LII pulse



- Cold soot from a quenched ethylene diffusion flame (custom-made soot generator): **mature soot**
- Laser-heating of soot particles
  - Nd-YAG laser : 1064 nm excitation wavelength
  - Beam diameter = 5 mm, Top-hat profile
  - Different laser fluences
- Wavelength-resolved extinction measurements
  - pristine particle (no laser irradiation)
  - at the LII peak
  - few seconds after the LII pulse



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Laser-induced incandescence (LII)

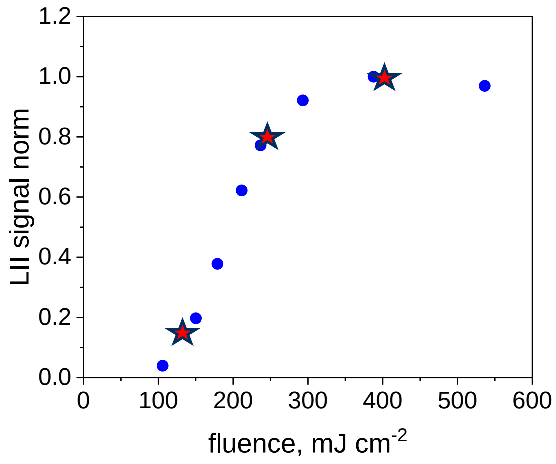
Good practice in LII measurements

→ look at the LII signal vs laser fluence curve!

Fluence curve depends on:

1. Particles properties
2. Experimental set-up (spatial and temporal laser profile, detection,…)

➡ ensure to be at a fluence equal to or greater than that required for the onset of the fluence plateau regime, while remaining below the fluence that produces significant sublimation



Three fluence investigated:

- ★ 130 mJ cm^{-2}: low fluence regime
- ★ 260 mJ cm^{-2}: moderate fluence
- ★ 400 mJ cm^{-2}: high fluence (close to sublimation)

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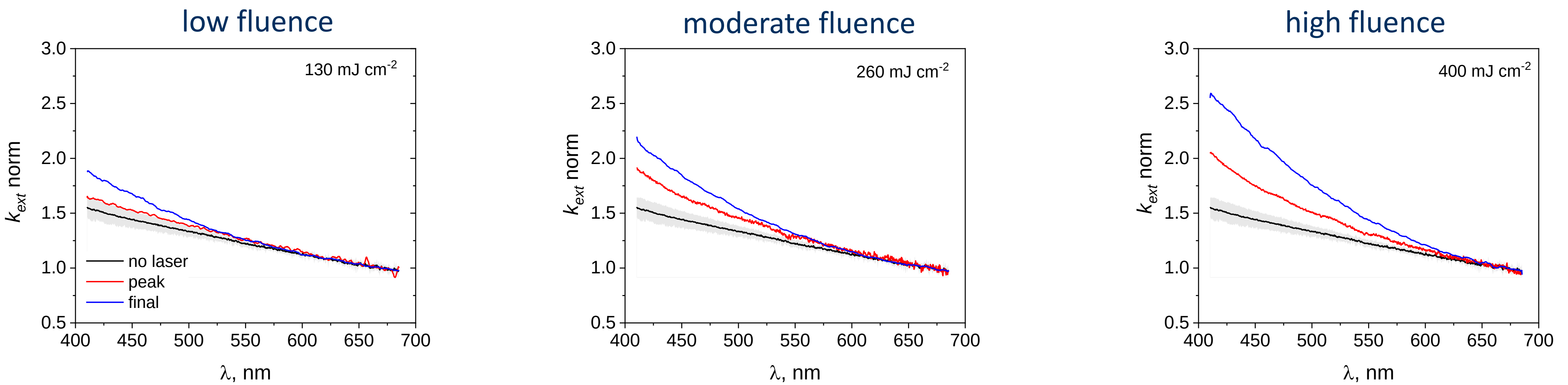
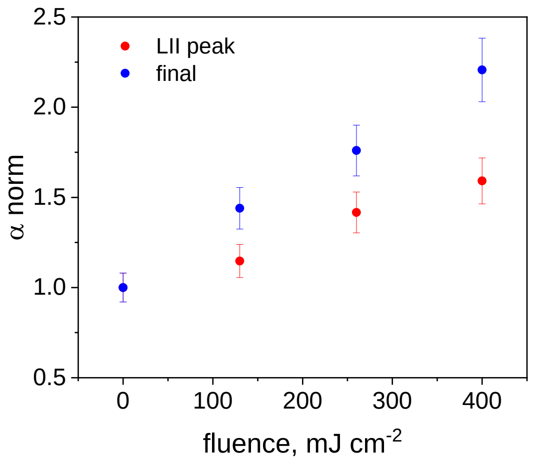
Wavelength-resolved extinction measurements

→Info on optical properties

- LII peak
- Final = few seconds after the LII pulse



In terms of dispersion coefficient, α



α ↑↑ with laser fluence!

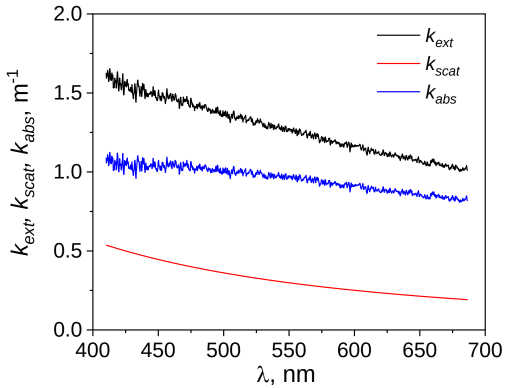
Scattering correction to extinction

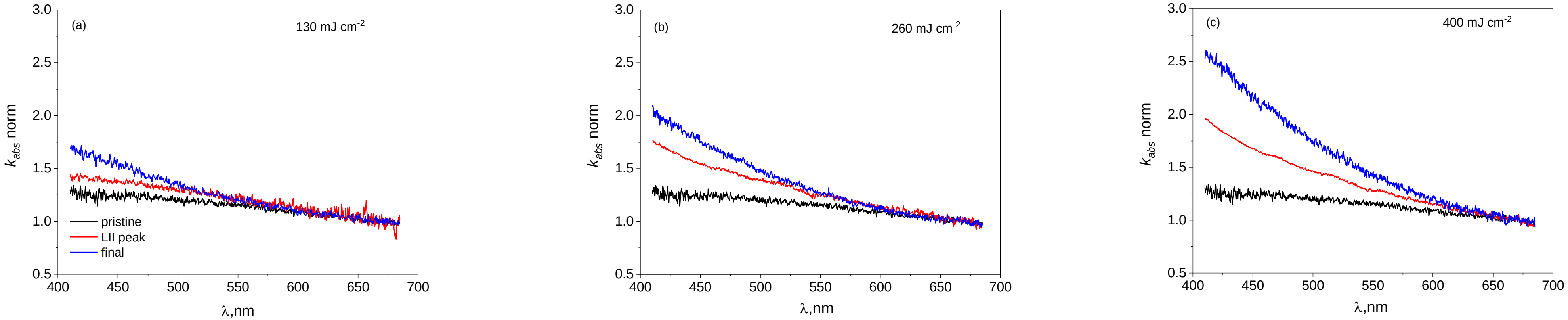
→ k_{ext} is obtained direclty from the transmissivity measurements

→ k_{scat} is evaluated assuming:

- Scattering Ångström Exponent (SAE) = 2
- Single Scattering Albedo at 480 nm from a previous work

→ $k_{abs} = k_{ext} - k_{scat}$





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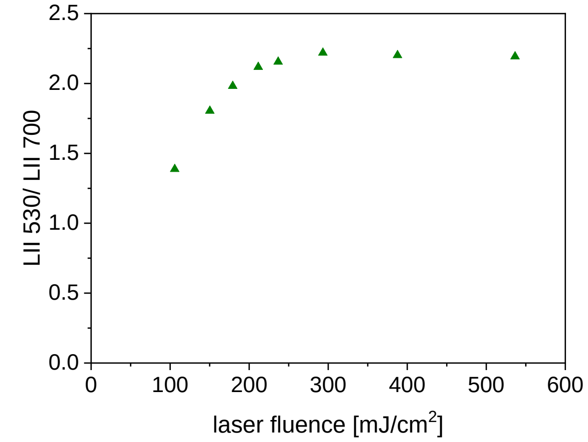
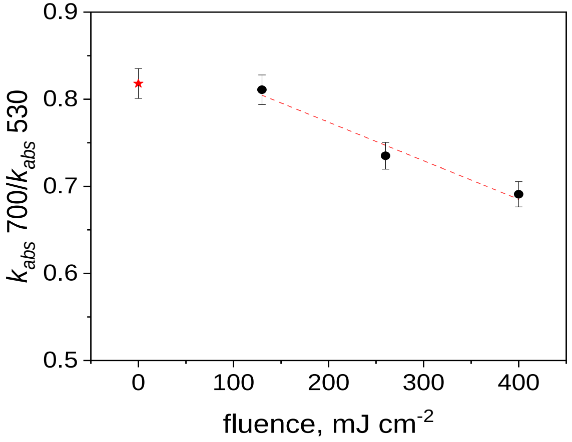
Impact on LII data interpretation

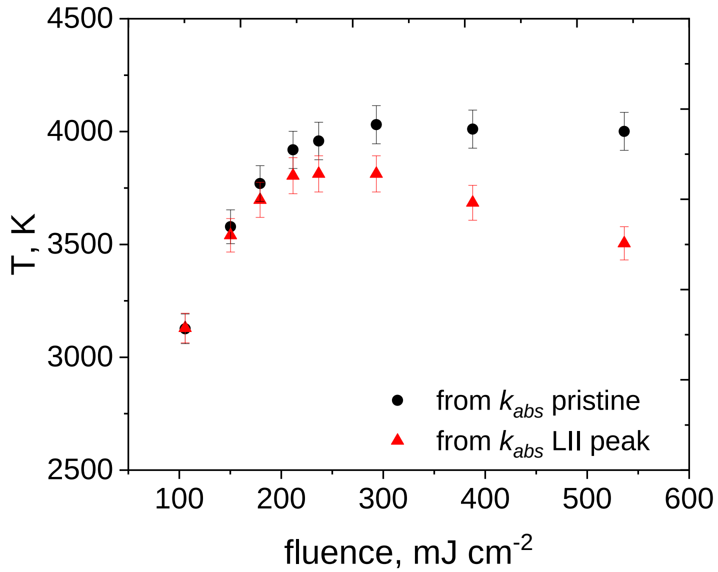
Particle temperature from 2C-LII

from a previous work





$$T = \frac{hc}{K_B}\left(\frac{1}{\lambda_2} - \frac{1}{\lambda_1}\right)\left[ln\left(\frac{S_1 k_{abs(\lambda 2)}}{S_2 k_{abs(\lambda 1)}}\right)\left(\frac{\lambda_1}{\lambda_2}\right)^5\right]^{-1}$$



evident reduction in the incandescence temperature is observed for fluences above 250 mJ cm^{-2}

Implication on soot volume fraction evaluation

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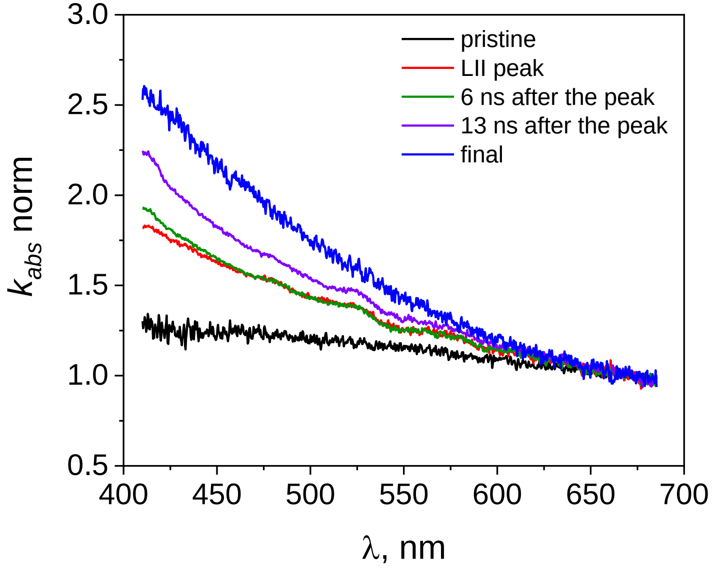
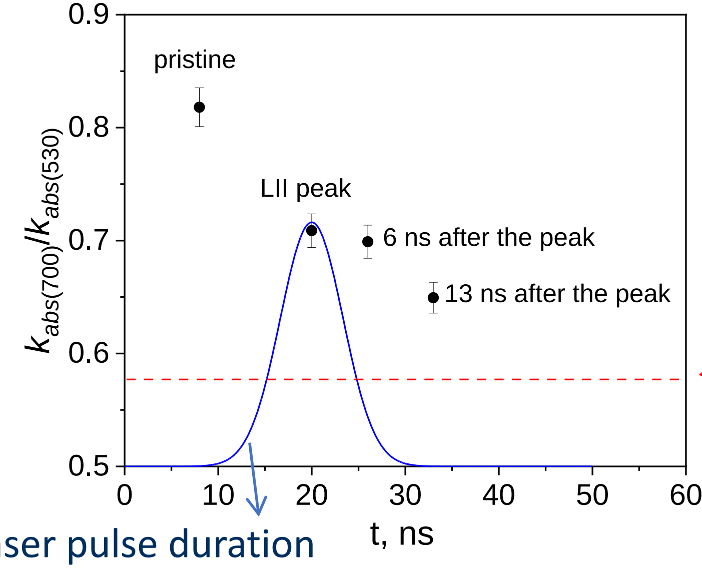
Absorption coefficient time evolution

Change of the absorption coefficient with time
@400 mJ cm^{-2}





laser pulse duration

k_{abs} ratio value at the final condition measurement

Significant change within the laser pulse

→ Possible implications on:

- Soot volume fraction
- Tire-LII → particle size
- Delayed LII detection
- Gated LII detection

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Take-home message

The knowledge of optical properties is paramount for:

- ✓ improving the understanding of soot formation pathways
- ✓ the interpretation of optical diagnostics

LII is a valid/robust diagnostic tool for studying practical system but:

- ✓ care has to be taken in the choice of the appropriate laser fluence for measurements
- ✓ the optical properties of the particles might change during the laser pulse depending on laser fluence and the nature of the particles
  - → the laser fluence threshold may vary according to the efficiency of the heating process

Emerging fuels (blended/additives/surrogates…) will be a new challenge

→ small particles, low concentration, different chemical composition

Wavelenght-resolved extinction measurements are a good option to gain information on optical properties

→ great advantage in the choice of LII detection wavelengths for particles temperature determination from pyrometry

Wavelenght-resolved extinction measurements might not be always practically feasible

→ we need more tools:

- ✓ LII itself can help (see Lang et al., Appl Phys B (2023) 129:147)
- ✓ Modelling
- ✓ Physical-chemical characterization of laser-irradiated particles
- ✓ …

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Thank you for your attention!