Can Black Carbon be defined by the absorption properties of laser-heated combustion generated nanoparticles ?

F. Migliorini, S. De Iuliis, R. Dondè, G. Zizak

francesca.migliorini@cnr.it

Introduction

Laser-induced incandescence (LII) is widely used for black carbon (BC) measurements

Effects of laser heating on carbonaceous nanoparticles:

- Changes in the particles internal structure
- Formation of new particles
- Permanent or reversible change in the optical properties
- …

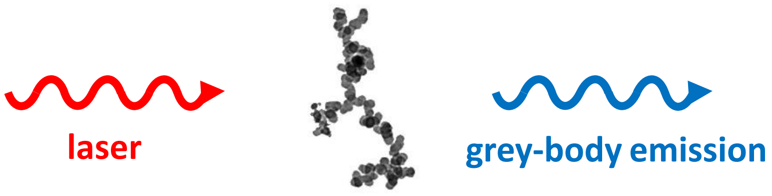
Affect laser-induced incandescence (LII) measurements and should be accounted for in the interpretation of the data

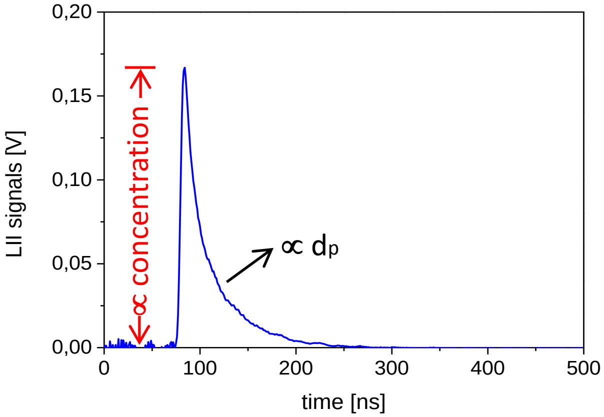
Objective:

Explore the spectral behavior of **carbonaceous nanoparticles absorption properties** after laser irradiation at different laser density energies

Laser-Induced Incandescence (LII)

LII involves heating particles with a high-power **pulsed laser** of several nanosecond and then measure of the incandescence emission signal





- Species selective → only CARBON
- High temporal and spatial resolution → on-line measurements
- High sensitivity and wide dynamic range → ng/m^3 – g/m^3

Laser-Induced Incandescence (LII)

LII signal strongly depends on the optical properties of the carbonaceous particles under investigation

$$T_s = T_g + \frac{6\pi E(m) R_0}{\lambda_{exc}\rho_{soot}\, c_{soot}}$$

T_g: gas temperature

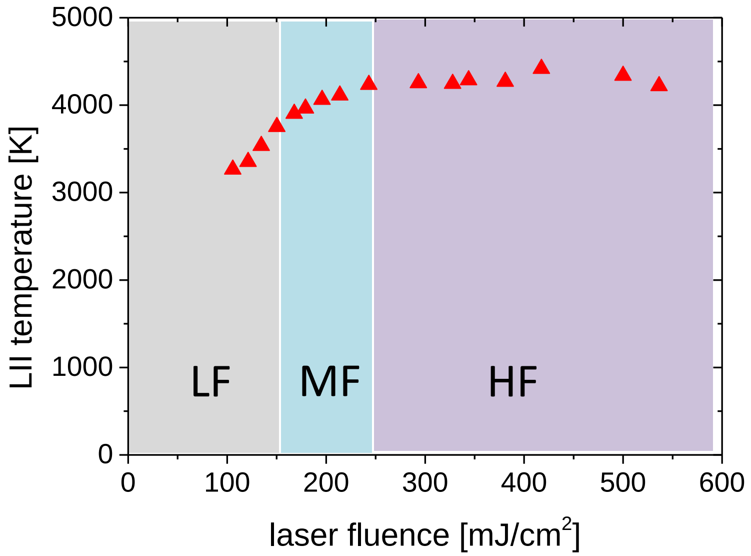
$E(m)$: refractive index absorption function

R_0: laser density energy (laser fluence)

λ_{exc}: excitation wavelength

ρ_{soot}: soot density

c_{soot}: soot specific heat capacity



LF: low fluence, **MF**: medium fluence, **HF**: high fluence

Methodology

1. *Generate a cold soot aerosol*

   - Home-made soot generator
   - Ethylene diffusion flame quenched by nitrogen flow

2. *Laser-heating of soot particles*

   - Nd-YAG laser : 1064 nm excitation wavelength
   - Beam diameter = 5 mm, Top-hat profile
   - Different laser fluences

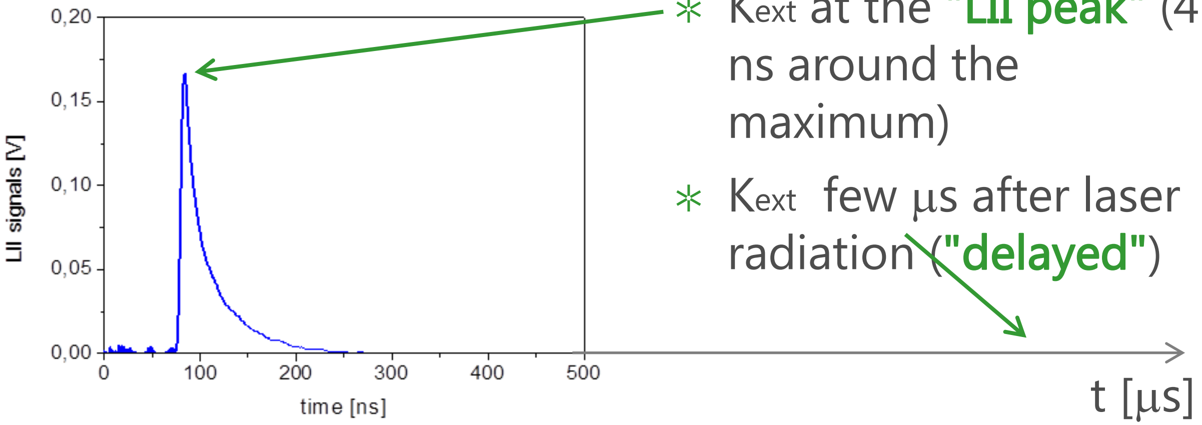
Methodology

3. *Evaluation of the spectral behavior of the absorption coefficient*

- Light attenuation measurements over a wide spectral range : 400- 680 nm

$$\frac{I}{I_0} = \exp(-K_{ext_\lambda} \mathrm{L})$$

- K_{ext} of cold and laser-heated carbonaceous nanoparticles

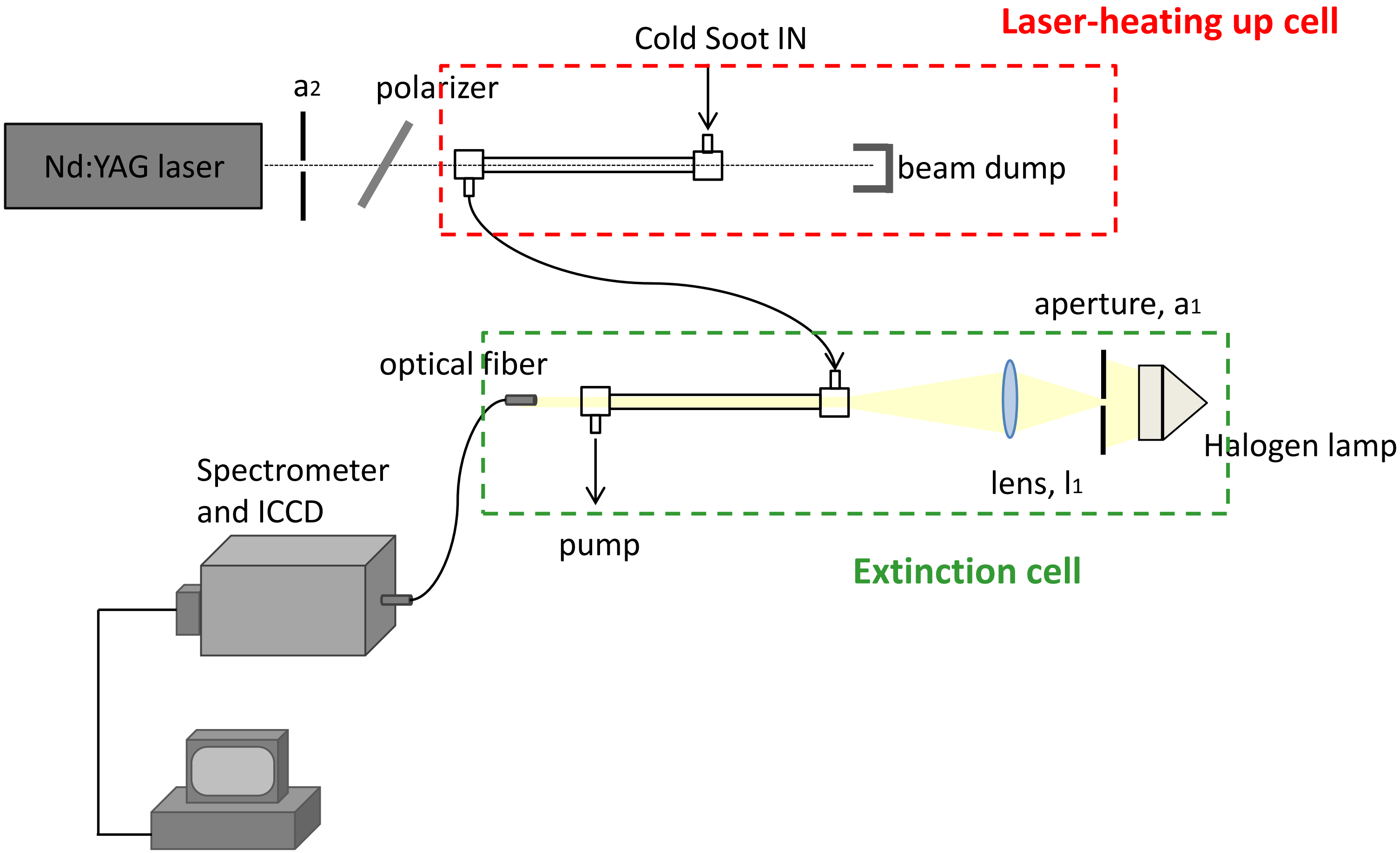


- K_{ext} at the **"LII peak"** (4 ns around the maximum)
- K_{ext} few µs after laser radiation (**"delayed"**)

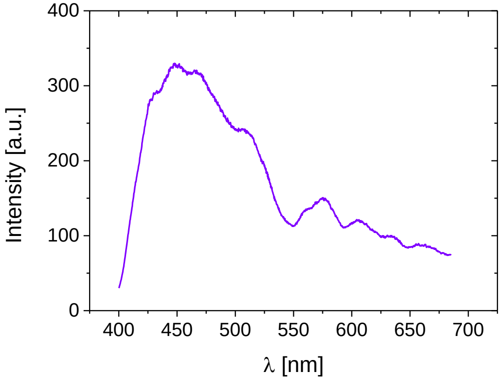
- If scattering is negligible:

$$E(m)_\lambda \propto K_{abs_\lambda}\lambda \simeq K_{ext_\lambda}\lambda$$

Experimental set-up – "Delayed" measurements

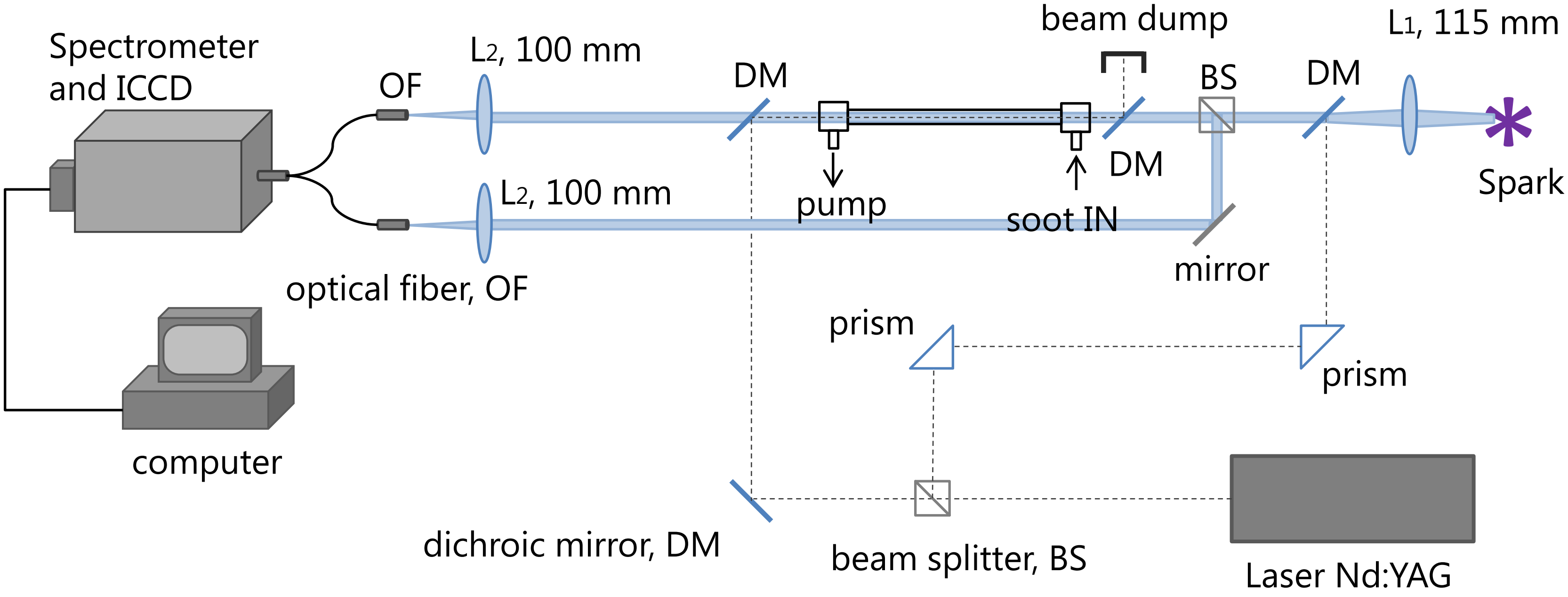


Experimental set-up – "LII peak" measurements



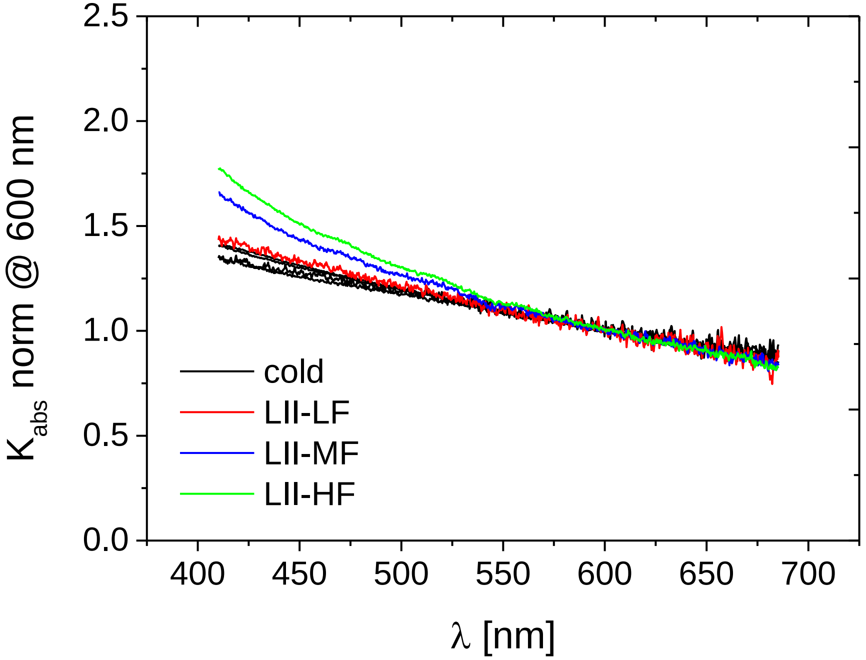
SPARK

- Broadband emission
- ≃30 ns duration

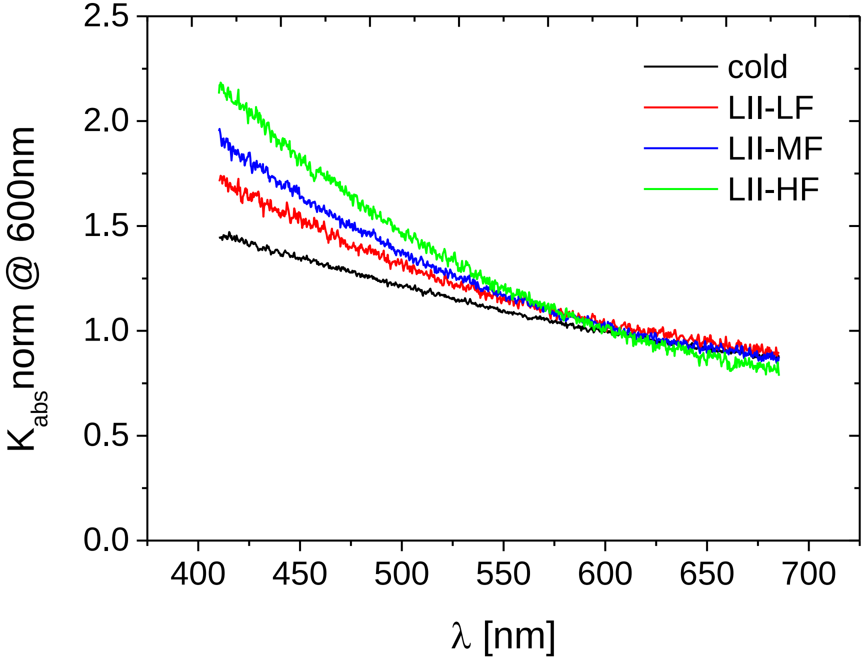


Results – "LII peak" vs "delayed" K_{abs}

LII peak

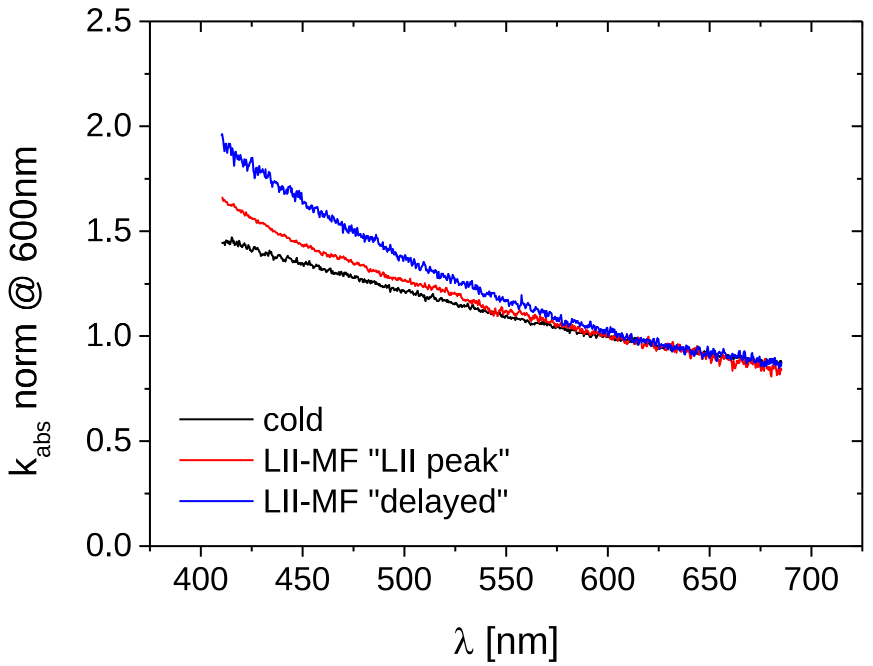


delayed

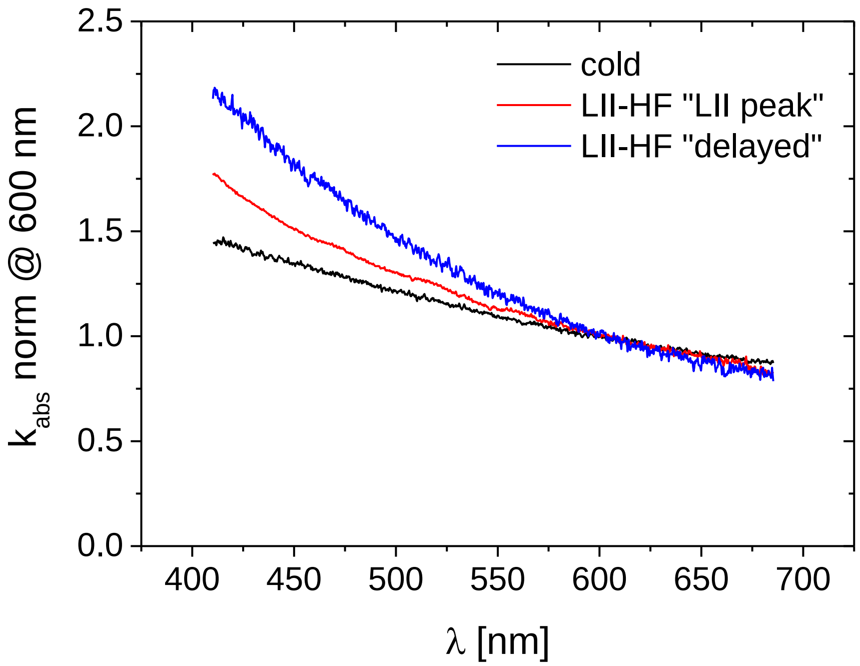


- K_{abs} is wavelength dependent
- Laser-heating of carbonaceous nanoparticles affects K_{abs}
- Laser fluence plays an important role

Results – Laser fluence effect



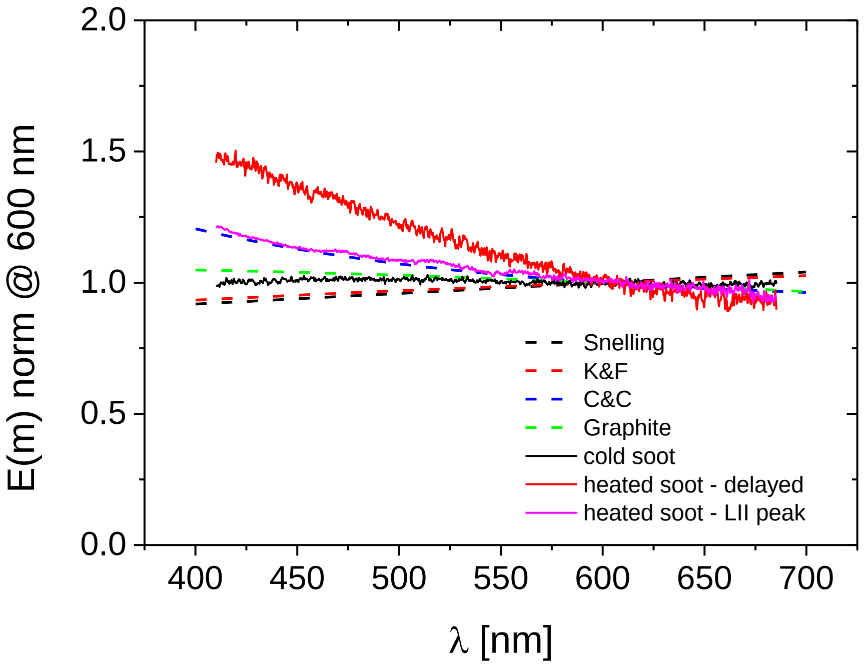
Medium fluence



High fluence

- The variation of K_{abs} starts already at the LII peak but becomes stronger after few μs

Results – E(m) wavelength dependence



- Laser-heating of carbonaceous nanoparticles affects the refractive index absorption function
- E(m) is wavelength dependent both for cold and heated particles
- E(m) of cold and heated carbonaceous nanoparticles show a different wavelength dependency

Results – Dispersion coefficient

The dispersion coefficient or Ångström exponent, α, is a useful quantity to assess the wavelength dependence of the "aerosol" optical properties.

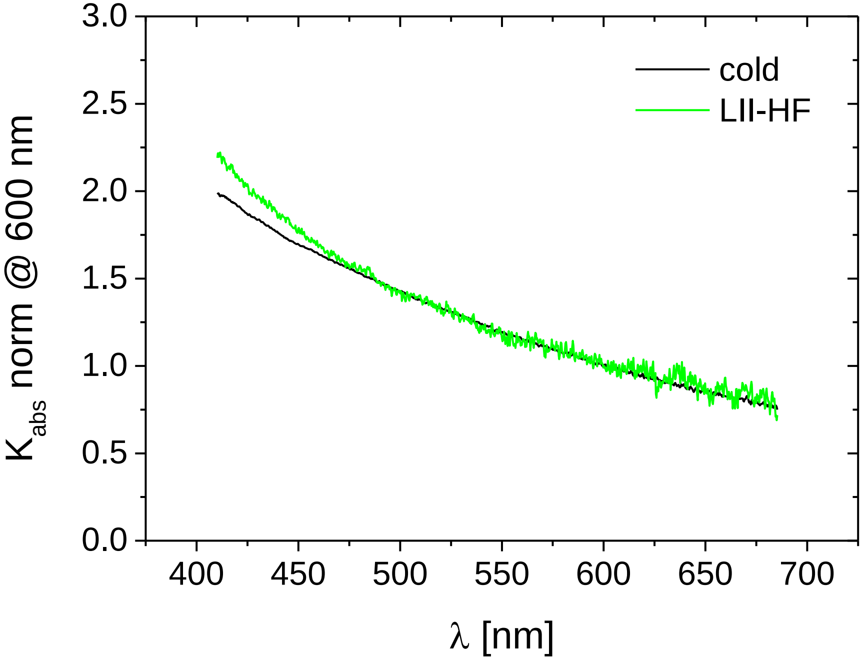
$$K_{abs,\lambda} = \frac{6\pi E(m) f_v}{\lambda} \propto \frac{1}{\lambda^{\alpha}} \quad \Rightarrow \quad E(m) \propto \frac{1}{\lambda^{\alpha-1}}$$

- α from nonlinear fitting of K_{abs} curves : 410-600 nm

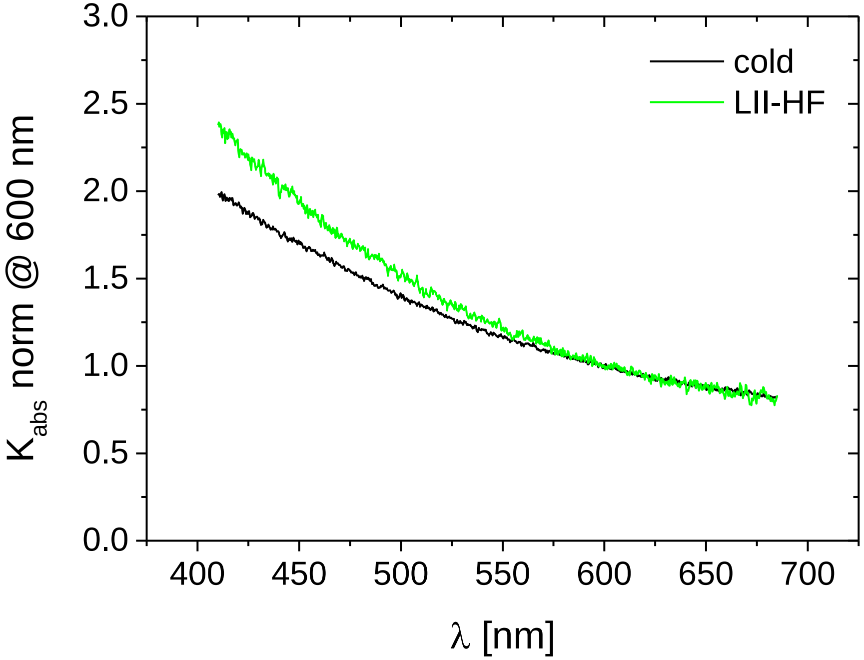
	LII peak [4 ns]	Delayed [µs]
cold	0.87	1.01
LII-LF	0.94	1.37
LII-MF	1.28	1.68
LII-HF	1.46	2.04

Results – Methane (preliminary)

LII peak



Delayed



- Laser-heating of soot particles affects K_{abs}
- For cold soot : dispersion coefficient higher than for ethylene (α= 1.8)
- For laser-heated soot: the dispersion coefficient is the same as for ethylene (α= 2)

Conclusions

- Extinction measurements have been performed to investigate the effect of laser heating on carbonaceous nanoparticles absorption properties.
- **Laser-heating** of carbonaceous nanoparticles affects the refractive index absorption function
- Such effect increases increasing laser fluence
- E(m) of cold and heated carbonaceous nanoparticles show a **different wavelength dependency**

Conclusions

- Carbonaceous nanoparticles generated by **Ethylene and Methane** and heated by a strong laser pulse (> 350 mJ/cm^2) show the **same spectral dependence of the absorption properties, once they have cooled down**.

⇩

1. **Best practice for LII measurements**

   choose wavelengths **>550 nm**

2. Room for a **working definition of BC**

   "any carbonaceous nanoparticle heated by a high laser density energy"

?

Future work

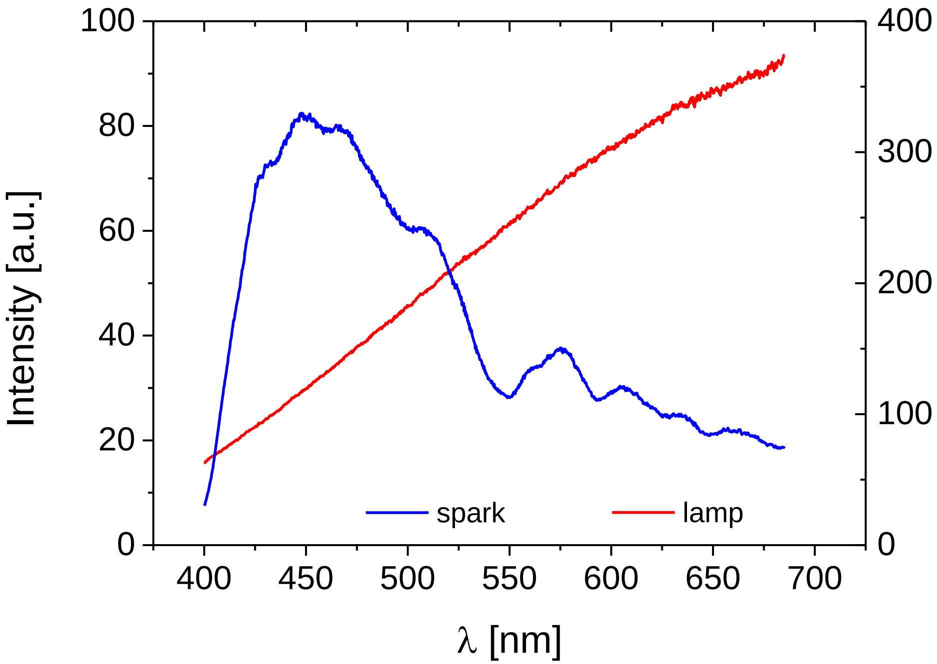
- Investigate "different types" of carbonaceous nanoparticles
- Morphological analysis of cold and heated carbonaceous nanoparticles via TEM
- Raman spectroscopy measurements to identify chemical bonds and vibrational frequencies responsible for the optical properties of the particles

Acknowledgment

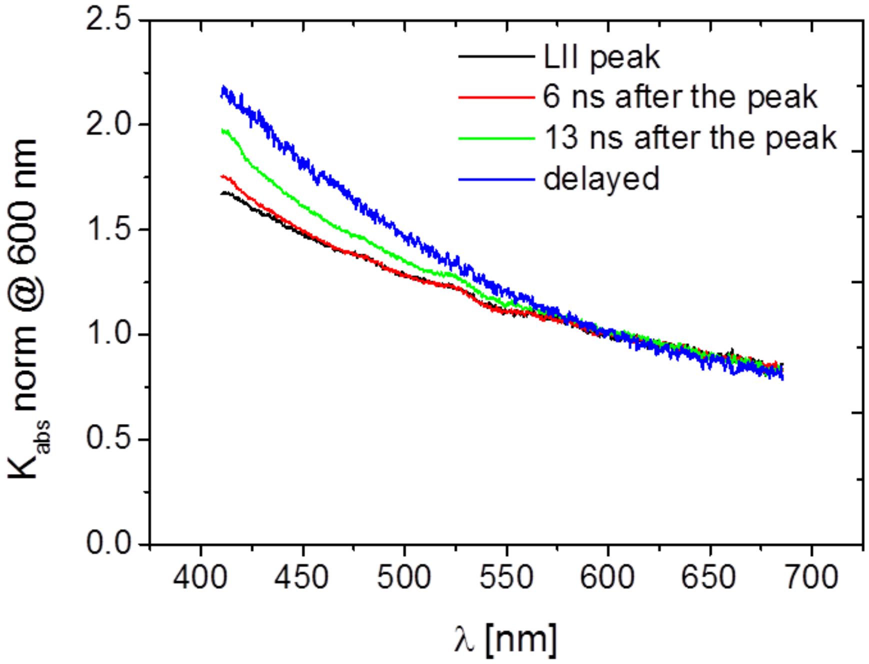
Financial support from the Ministero dello Sviluppo Economico (MSE)

Thank you all for your attention !

Light sources



K_{abs} evolution with time



K_{abs} cold soot