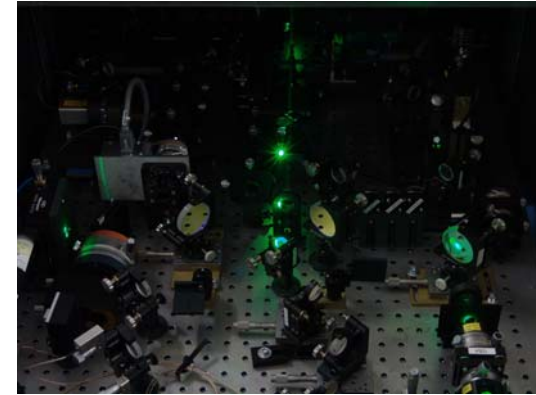


LII Workshop Session

Experimental Issues

Aug. 04, 2006, 11:25 – 12:25, 13:25 – 14:55



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Experiments performed

laminar diff. laminar diff. laminar diff.	isooctane defined aliphatic/aromatic mix palm oil	1 bar 1 bar 1 bar	0D TiRe LII/2D simultaneously	hot
laminar prem.	C ₂ H ₄ /air	1-3 bar	0D TiRe LII	hot
laminar diff. or laminar prem.	CH ₄ (/air) C ₂ H ₂ (/air) kerosene spray	0.15-0.4 bar 1 bar	2D	hot
laminar prem.	C ₂ H ₄ /air	≥ 1 bar	0D TiRe LII and 2D	hot
laminar diff. or laminar prem.	C ₂ H ₄ (/air)	1 bar	0D TiRe LII	hot
laminar diff.	CH ₄ C ₂ H ₄	1 bar	2D + 2D TiRe	hot
laminar prem.	C ₂ H ₄ /air	1 bar	0D	hot
laminar prem.	C ₂ H ₂ /air	1 bar	1D + TiRe LII	hot
laminar diff.	C ₂ H ₄	1 bar	0D TiRe 2C-LII	hot



Experiments performed

laminar diff.	CH ₄	1-80 bar	0D TiRe 2C-LII	hot
quenched laminar diff.	C ₂ H ₄	1 bar	0D TiRe 2C-LII	moderate
sampled from laminar diff.	C ₂ H ₄ flame, soot diluted in He, Ne, Ar, Kr, Xe, N ₂ , CO ₂ , SF ₆	1 bar	0D TiRe 2C-LII	ambient
turbulent	diesel and similar (engine)	High	0D TiRe LII	hot
turbulent	diesel (engine)	up to 75 bar	0D TiRe LII	hot
turbulent	diesel (engine)	50-70 bar	0D TiRe LII/2D simultaneously	hot
	Soot generator from C ₃ O ₂	1 bar Ar		ambient - hot
after flame		1 bar	2D	moderate
carbon black		< 10 ⁻³ mbar	0D TiRe LII/2D	ambient
carbon black		1-30 bar	0D TiRe LII/2D simultaneously	ambient
Fe	Fe nanoparticle reactor	0.5 bar	0D TiRe LII	349 K



Laser Attenuation

In most cases, full laser output is too strong for LII experiment (2D and TiRe-LII)

- Polarizer + thin $\lambda/2$ plate (no change of beam profile nor direction)
- Rotating Glan-Thompson polarizer (change of beam direction? Makes linearly polarized incoming light desirable that is not provided by all frequency doubling crystals – case reported is using 532 nm)
- Set of ND filters (risk of overheating, beam misalignment)
- Angle dependent, reflecting attenuator plate (change of profile/direction? Variability?)
- Laser control unit (?)
- Q-Switch timing
- Lamp Joules (change of focus position in dimension of centimeters)
- Variable beam expansion



Measurement of spatial laser energy distribution

Required for determination of laser fluence (2D and TiRe-LII)

- Scanning a 10 μ m pinhole with a photodiode/energy detector across the beam/sheet (1D)
- Scanning a knife edge through the laser (1D)
- Burn pattern with polaroid paper (unknown, nonlinear sensitivity)
- Beam profiler (plus telescope/lens if sheet is studied)
advantage: shot-to-shot 2D

- Should be shown in publications!
- If the profile is not uniform, discuss the impact on the results presented
- What constitutes uniformity?



Measurement of temporal laser energy distribution

Essentially required for calculating the sublimation term (TiRe-LII)
For gated LII only important if strongly different from standard (5-7 ns)

- Fast (?) photodiode + acquisition system (any reflection suitable)
- 1 ns photodiode + 1 GHz oscilloscope (any reflection suitable)
- ≤ 1 ns photodetector + 20 GS/s oscilloscope (any reflection suitable)
- Assumption: Gaussian (duration?)



Position of monitoring the laser energy

Required for calculating particle heating term (TiRe-LII)
For gated LII important for intensity-to-fluence diagram, either determined once or for each experiment having a changed design

- Powermeter, flame radiation subtracted
- Photodiode calibrated with pyrodetector → single shot info into PC
- GENTEC PS-V-103 powermeter
- ICCD
- Behind the flame
- In the position of the flame before the experiment (only suitable if laser power constant and no changes during the experiment are required)
- Before the flame
- Before and behind measurement object

Best position depending on dimension and concentration of sooting object – concern: calibration of powermeter



Devices of homogenization of laser

Should be as homogeneous as possible → heating all particles in measurement volume similarly

- Clipping most homogeneous parts out of a expanded sheet by an aperture (clipping in 2D) → dimension of sheet thickness will be Gaussian in focus!
- Aperture selects homogeneous part of the beam – relay imaged onto burner center



Wavelengths

Excitation / nm	Detection / nm	Interferences
1064	405 ± 5 / 650 ± 5 (2C) 441,6 ± 5 / 650 ± 25 (2C)	none
1064	400 / 700 (2C)	
1064 (+532 for PAH)	Broadband vis or RG630	
532		PAH, C ₂
1064	400-650 (resolved)	C ₂ (?)
1064	550 / 694 (2C)	
1064	460 / 650	
1064	Broadband vis 500 ± 12.5 / 700 ± 12.5	
1064, 532	397±19 or 445±30 / 782±6 (2C) 445±30 / 750±25 (2C)	PAH?, C ₂ ?
532	405	PAH
532	450 ± 20 / 650 ± 12.5	No PAH/C ₂ expected?
532	410	Particle size effects?
		LII from ... ?



Timing and synchronization

Delayed and long gate favors big particles in measurement volume in 2D soot concentration measurements (according to Lit.)

- Gates applied: 10 ns, 20 ns, 25 ns, 40 ns (prompt), 40 ns delayed
- Synchro using Q-Switch delay out
- Synchro using delay generators
- Gate opening at LII maximum
- Peak signal used from time trace with 2 ns resolution
- TiRe with analysis starting from 10 ns after peak of laser pulse

- How is “prompt” defined?
LII peaks later (... ns) than laser!

- In practice: detection of Rayleigh scattering for zero definition



Detection devices (2D)

- 12 bit ICCD for 2D f_v
- ICCD + Spectrometer for 1D spatially + spectrally resolved signal
- 12 bit ICCD double frame for 2D f_v + background
- Streak camera for 1D spatial d_p , min. exposure 5 ps
- ICCD temporal scan with 5 ns gate (stable flame required) for f_v and d_p (lower laser fluence for the latter)



Detection devices (0D)

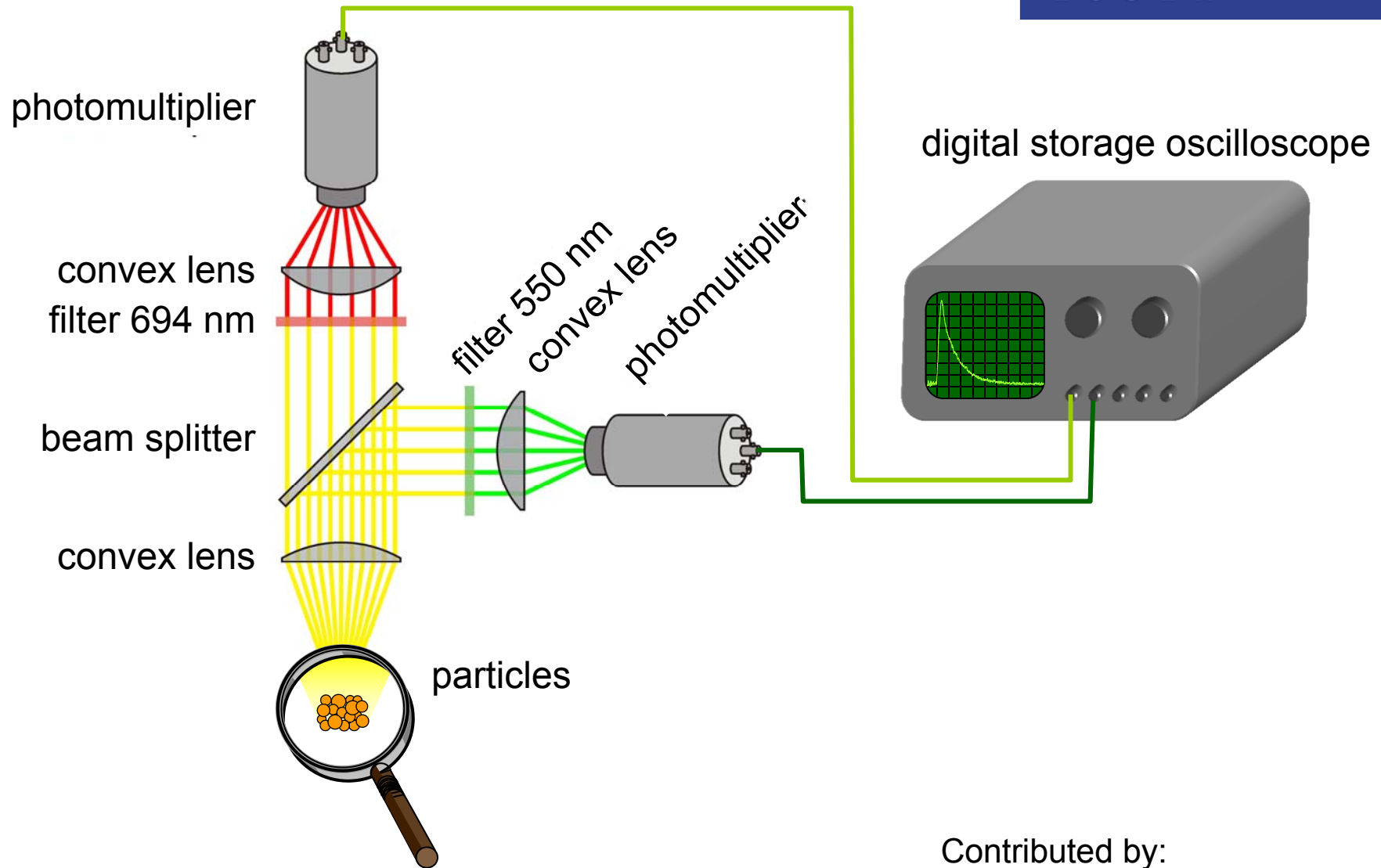
- PMT: Hamamatsu H7710, rise time 1.4 ns
- PMT: Hamamatsu R928, rise time 2.2 ns
- PMT with integrated amp:
SMT MEA1030 V8DA + Hamamatsu R7400U-04, rise time 0.78 ns
- PMT (type?) with rise time <0.4 ns
- PMTs: Hamamatsu H5783-03 bialkali and R7400U-50 multialkali, rise time ~ 1 ns
- PMTs: Hamamatsu R1924A bialkali and R5900U-20 multialkali, rise time ~ 1.5 ns

- Oscilloscope 300 MHz, 2.5 GS/s
- Oscilloscope 500 MHz
- Oscilloscope 1 GHz, 2 GS/s (0.5 ns)
- Oscilloscope 2 GHz, 20 GS/s

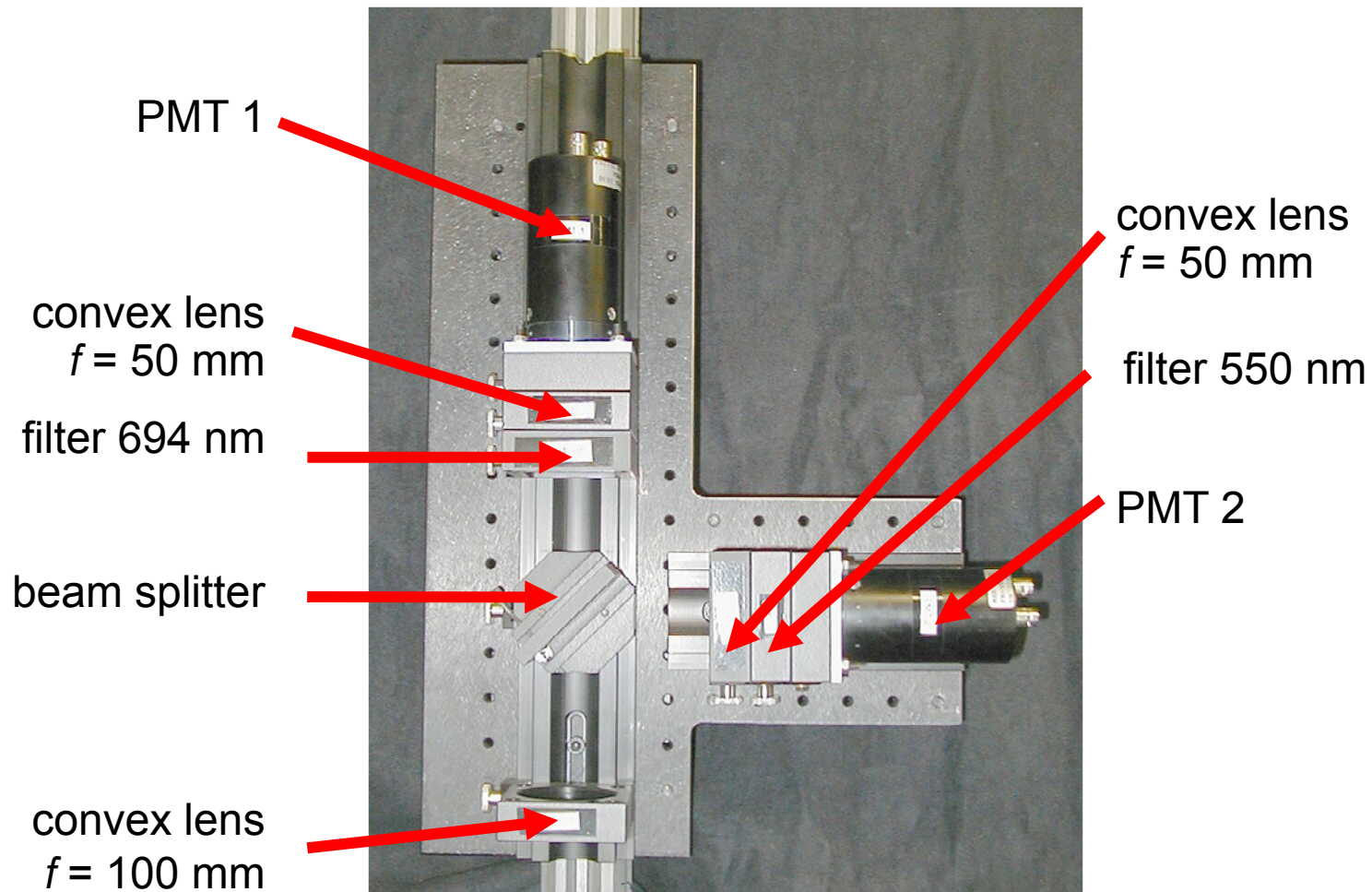
- 50/50 beam splitter for 2C experiments
- Dichroic beam splitter for 2C experiments



Two-color detection system



Two-color detection system



Detector response function

- Monitor scattered laser radiation
- Triggering from single photon events and averaging over a large number



Minimal detected soot concentrations

- 2 ppm in 1 mm x 0.8 mm cyl. measurement volume (2D)
self calibrated TiRe LII
- < 1 ppb in 1 mm³ (2D)
CRDS
- 1 ppb within ± 1.5 mm (“0D”)
extinction calibration (laminar flame)
- 0.1 ppm
extinction calibration
- 2 primary particles in vacuum (and close to one!) within ± 1.5 mm
no conduction \rightarrow very long τ
- 50 ppb with lots of averaging in laminar flames (2D)
extinction calibration
- below 0.010 ppt (<20 ng/m³) in 1550 mm³ (0D?)



Estimated accuracy

- Approx. 2 nm @ 20 nm, relative error increasing for smaller d_p
- Approx. 10 % in raw signal for f_v and in decay for d_p
- Approx. 10 – 15 % for d_p
- Approx. 30 % absolute for f_v due to photometric calibration, $E(m)$...
- Approx. 20 % mainly due to $E(m)$

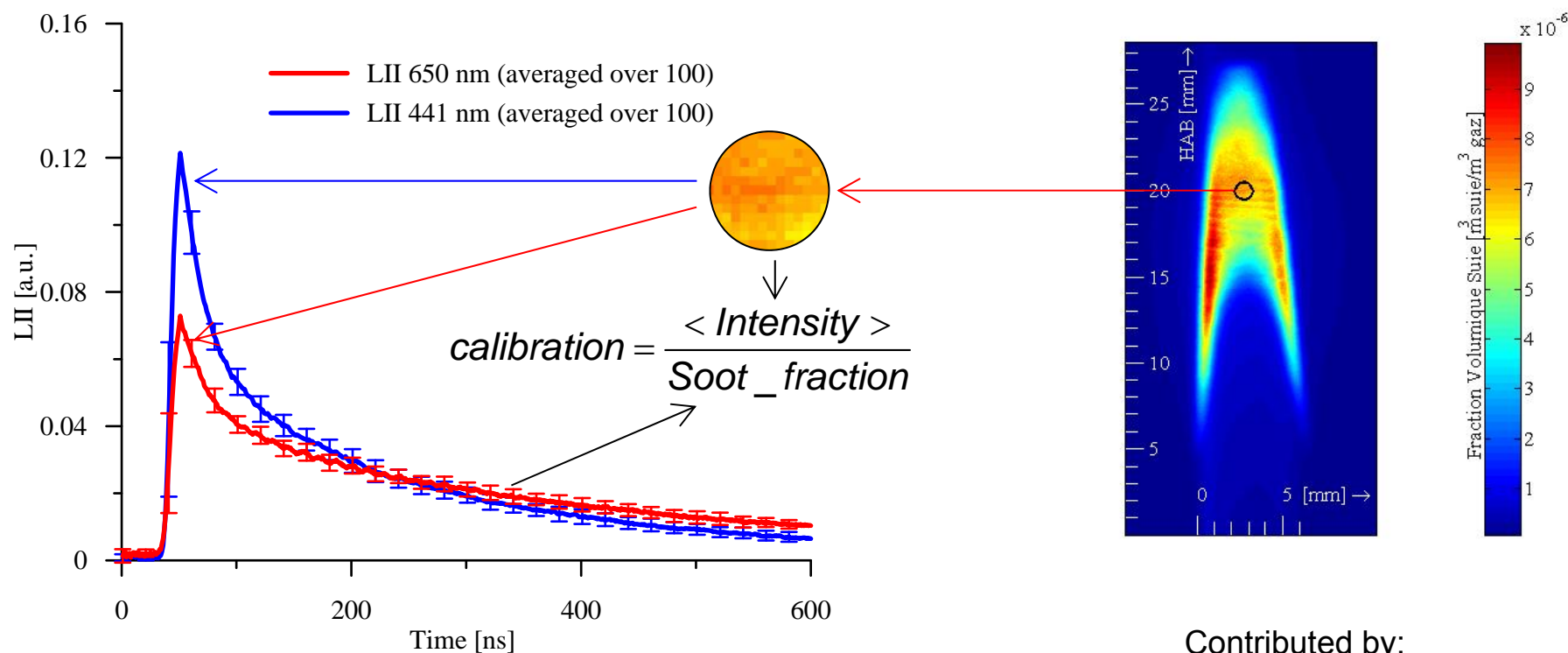


Calibration technique (0D)

- 2D f_v measurement calibrated by 0D self calibration technique at moderate laser fluence for both calibration object: Osram lamp at defined T, chopper wheel performed in position of measurement
low laser fluence used for 0D and simultaneous 2D experiment
- 0D and 2D f_v measurement calibrated by 0D self calibration technique calibration object: calibrated hot tungsten strip lamp
- 0D f_v and d_p measurement calibrated by 0D self calibration technique calibration object: spectral radiance calibrated tungsten filament bulb, quartz halogen bulb, or integrating sphere



Simultaneous Time-Resolved / 2D LII



Contributed by:
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 C. Mounaïm-Rousselle
 University of Orléans



Calibration technique (1 or 2D)

- f_v measurement calibrated by extinction (633 nm) in the same flame (laminar premixed pressurized)
same optical path through the flame should be used for ext. and LII
extinction should temporally be separated from LII process
plane parallel windows might induce etalon effects
- 2D f_v measurement calibrated by CRDS (low soot concentrations) or extinction in several heights in the flame (temporally stable flames)
best use 1064 nm because even 633 nm might be influenced by PAH
- 2D f_v measurement calibrated by extinction (532 nm), same beam as LII, attenuated by factor of 100, same flame (laminar diffusion)
extinction should exceed 10 %



Calibration technique (1 or 2D)

- 2D f_v measurement calibrated by extinction (532 nm), in same flame (laminar premixed pressurized) or using laminar premixed atmospheric calibration burner in position of expt.
pulsed extinction laser has to be sufficiently attenuated!
- 0D f_v measurement calibrated by line of sight extinction (633 nm) in the same flame (turbulent) using homogeneous soot distribution as initial guess



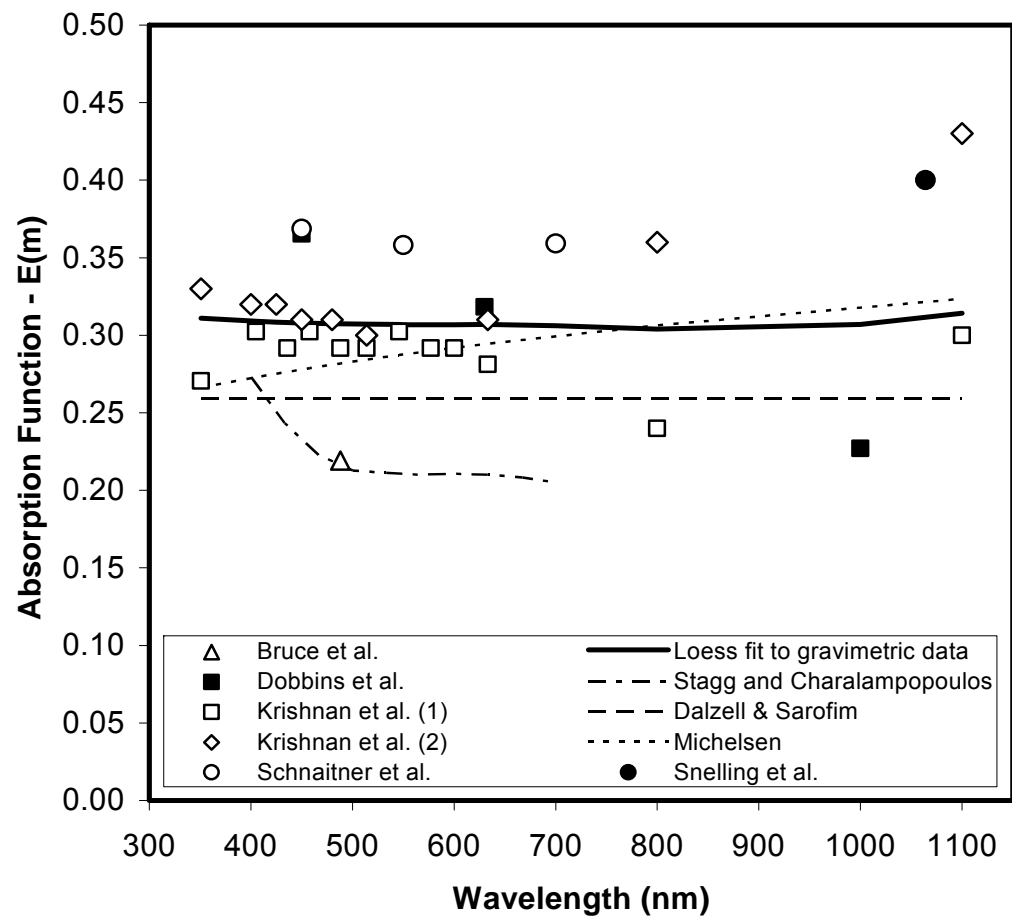
Optical soot properties used ...

- “Dalzell, Sarofim” for extinction based calibration @ ? nm
- $E(m) = 0.4$ @ 633 nm according to “Smallwood et al.”
- $E(m) = 0.4$ @ 1064 nm according to “Smallwood et al.”
- $E(m) = 0.4$ as “accepted value”, trying own measurements
- $E(m) = 0.232 + 1.2546 \times 10^{-4} \times \lambda(\text{nm})$ @ 400-1064 nm, according to “Krishnan et al.”
- $m = 1.57 - 0.56i$ @ ? nm
- $m = 1.75 - 0.58i$ @ 633 nm according to “Chang”
- $m = 1.60 - 0.59i$ @ 532 nm according to “Charalampopoulos”
- $K(\text{ext}) = 10$ @ 532 nm, to match value in numerical simulation

- Reasons to prefer the one or the other?



Absorption function determined for flame and Diesel soot over the UV-VIS-NIR spectrum



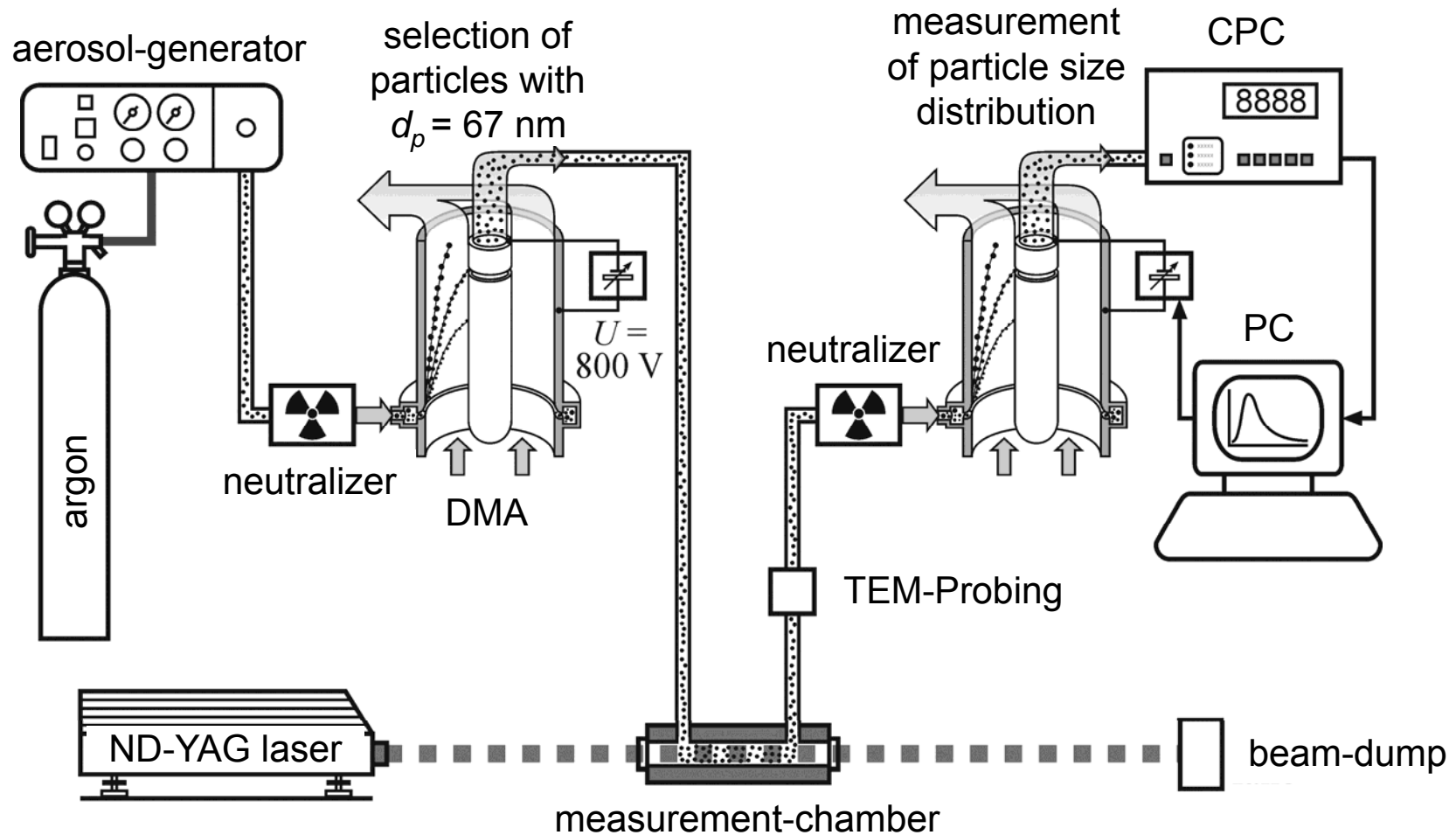
Additional experiments/information available

- Mass spectroscopic particle sizes in McKenna (DLR)
- 2C LII gives access to relative $E(m)$ at both λ (Uni Lille)
- Mass spectroscopic study of PAH adsorbed on soot particles (Uni Lille)
- Influence of laser fluence on particle morphology or aggregate structure (→ Vander Wal, Uni Duisburg) *
- Influence of laser fluence on aggregates/agglomerates (Uni Cranfield)
- Influence of laser fluence on soot volume fraction (NRC/Columbian)
- LII at high temporal resolution (→ Sandia) - extension desired *
- Defined in-situ soot generation: C3O2 pyrolysis (H-free soot!) – **availability to LII researchers?**
- Accomodation coefficient from shock tube experiments (Uni Duisburg) – **commonly valid?**
- Accomodation coefficient from noble gas (+others) experiments (NRC)

- *** see below**



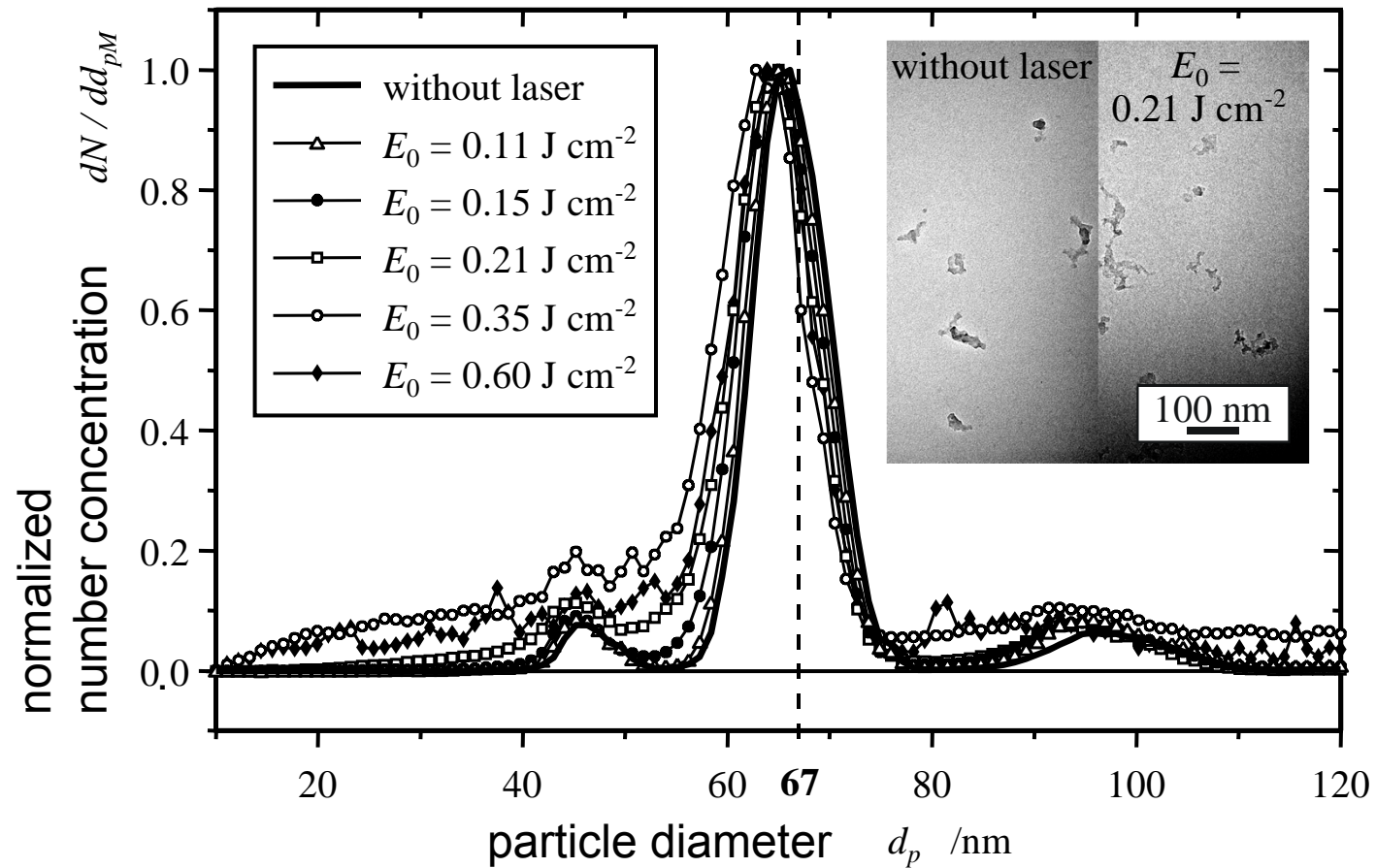
Influence of laser fluence on soot-particle size and morphology



Contributed by:
B.F. Kock, C. Schulz, P. Roth
IVG, University Duisburg-Essen



Influence of laser fluence on soot-particle size and morphology

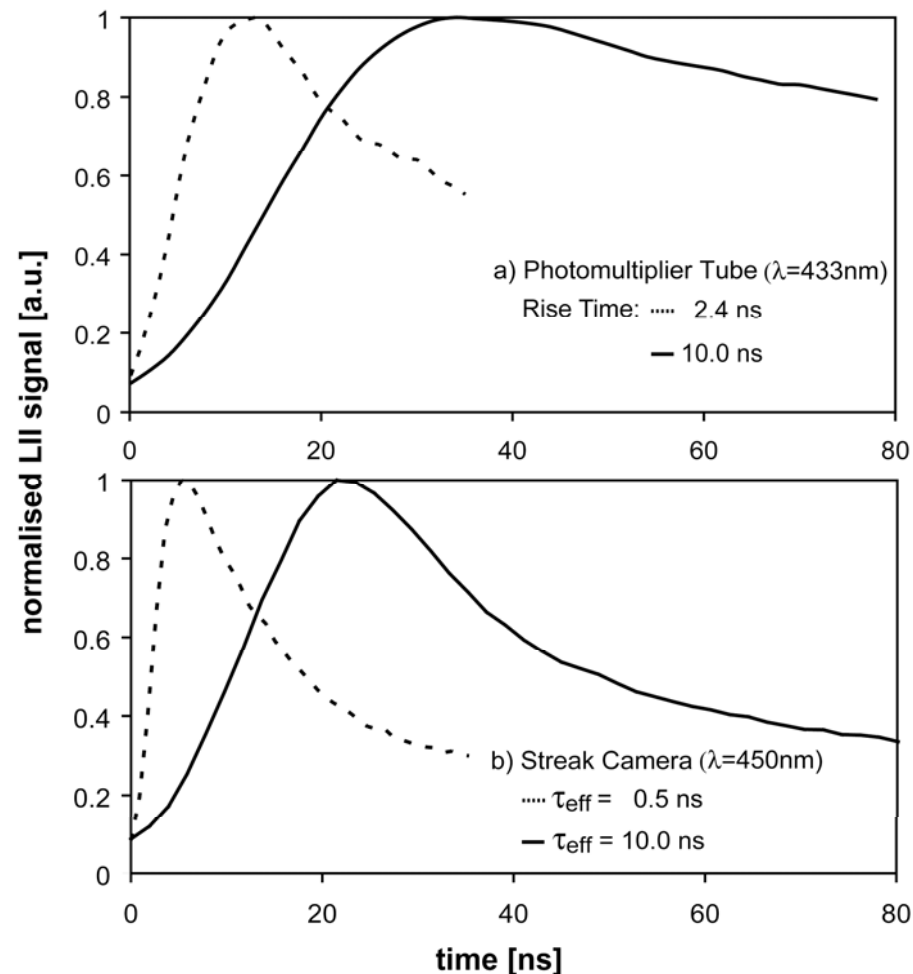


Fast LII - LII Signal Evolution: Influence of Temporal Response



Detection device:	Streak Camera	PMT/Oscilloscope
Pressure:	Atmospheric pressure conditions	
Size distribution:	Log-normal; $d_m=15.2\text{nm}$, $\sigma=0.34$	
Detection wavelength:	$\lambda=450\text{nm}$	$\lambda=433\text{nm}$
Exposure time/Rise time	0.5ns; 10ns	2.4ns; 10ns

➔ An insufficient temporal response affects the normalised time-resolved LII signal evolution.

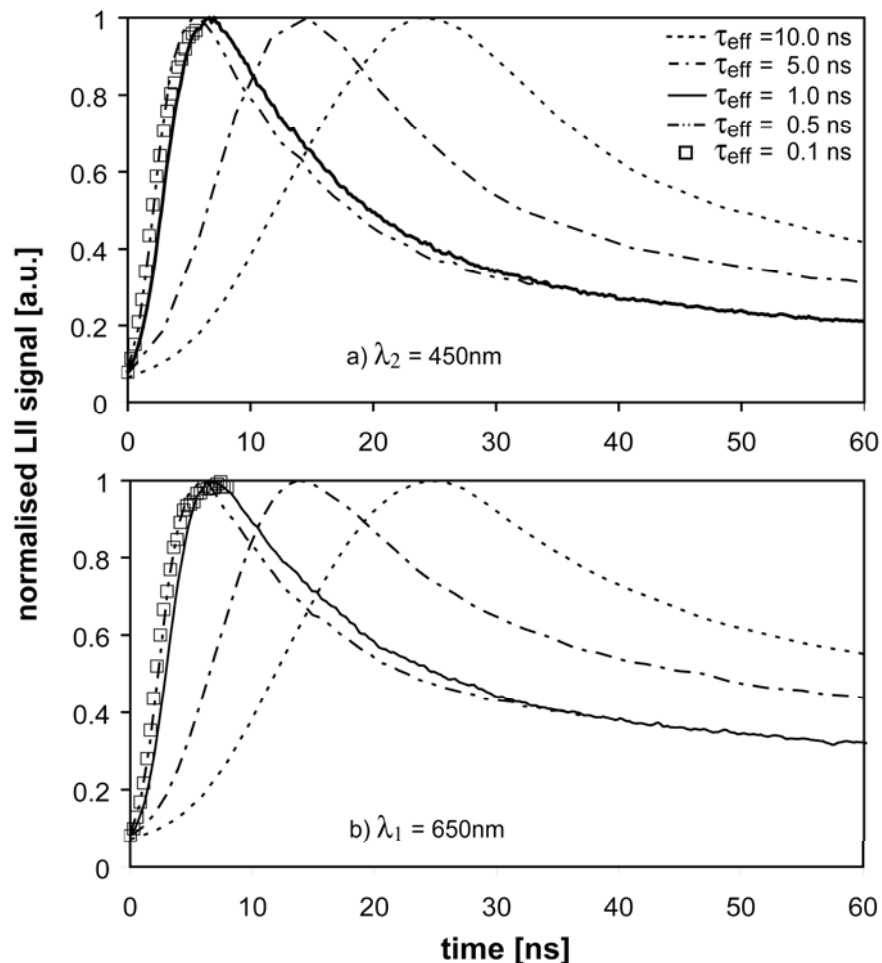


Contributed by:
M. Charwath, R. Suntz, H. Bockhorn
University of Karlsruhe (TH)






Experimental Results



Experiments:

Measured time-resolved LII signals of a known particle size distribution ($d_m=15.2\text{nm}$, $\sigma=0.34$). The signals are detected with different exposure times of the streak camera.

 An increased exposure time results in slower signal rise and fall rates.

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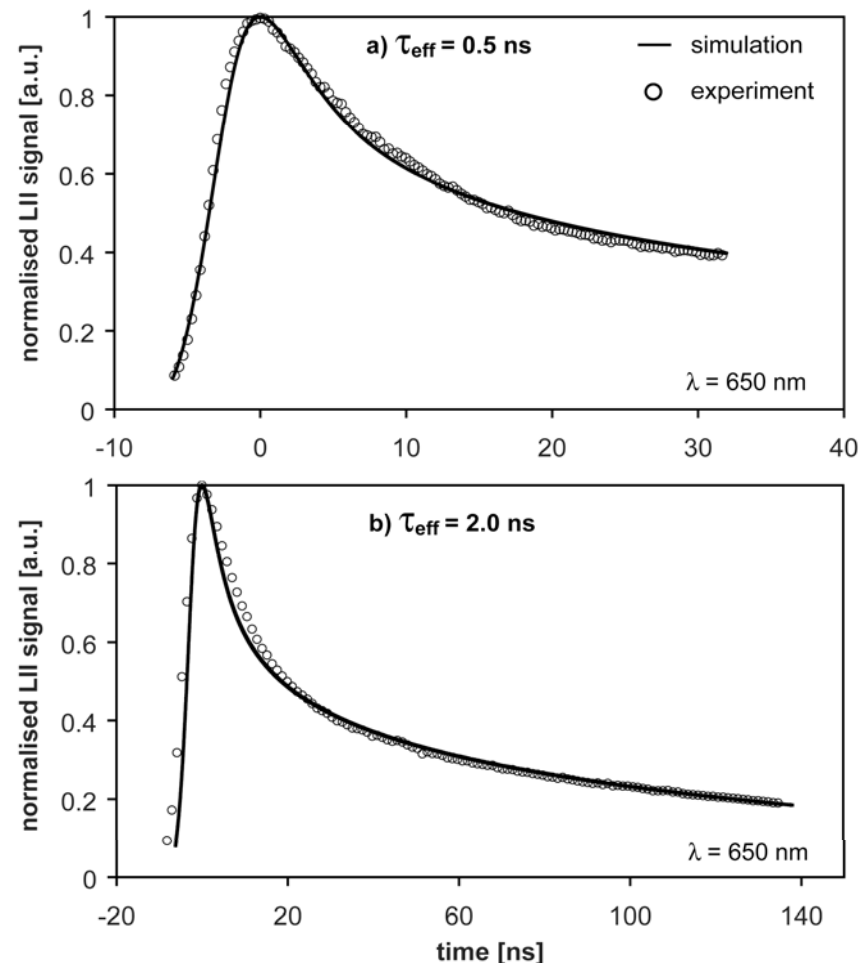
Experimental Results

Exposure time:	0.5ns	2.0ns
Accommodation coefficients:	$\alpha=0.28$ $\alpha_v=0.9$ $n=0.4$	$\alpha=0.32$ $\alpha_v=0.4$ $n=0.46$

➔ Significant influence of an inappropriate temporal response on the determination of accommodation parameters from TIRE-LII signals applying LII-models including particle heating as well as particle cooling. Only slightly affecting particle size if data evaluation starts some ns after maximum.

Contributed by:

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Burning issues – next experiments

- Ideas for 3D calibration in turbulent flames:
Multi-laser light extinction for “single shot” tomography (might suffer from very tiny soot structures typically present in turbulent flames)
- LII calibration for high pressure applications (f_v)
- 2D 2C LII for determination of T of laser-heated particles across the laser sheet
- Mass spectroscopic study of species vaporizing from particle surface upon pulsed laser radiation
- Spectrally resolved TiRe LII for (spectrally) comparing carbon/soot incandescence with black body
- Simultaneous experiments (adding info on T, gas composition ...)
- New alternative d_p and f_v diagnostics?
maybe comparison with concentration measurements by digital camera pyrometer available next workshop

