LII Workshop Session
Experimental Issues


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German Aerospace Center (DLR), Stuttgart, Germany
## Experiments performed

<table>
<thead>
<tr>
<th>Experiment Type</th>
<th>Fuel Type</th>
<th>Pressure</th>
<th>Technique</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laminar diff.</td>
<td>Isooctane</td>
<td>1 bar</td>
<td>0D TiRe LII/2D</td>
<td>Simultaneous hot</td>
</tr>
<tr>
<td>Laminar prem.</td>
<td>C₂H₄/air</td>
<td>1-3 bar</td>
<td>0D TiRe LII</td>
<td>Hot</td>
</tr>
<tr>
<td>Laminar diff. or laminar prem.</td>
<td>CH₄/air, C₂H₂/air, Kerosene spray</td>
<td>0.15-0.4 bar</td>
<td>2D</td>
<td>Hot</td>
</tr>
<tr>
<td>Laminar prem.</td>
<td>C₂H₄/air</td>
<td>≥ 1 bar</td>
<td>0D TiRe LII and 2D</td>
<td>Hot</td>
</tr>
<tr>
<td>Laminar diff. or laminar prem.</td>
<td>C₂H₄/air</td>
<td>1 bar</td>
<td>0D TiRe LII</td>
<td>Hot</td>
</tr>
<tr>
<td>Laminar diff.</td>
<td>CH₄, C₂H₄</td>
<td>1 bar</td>
<td>2D + 2D TiRe</td>
<td>Hot</td>
</tr>
<tr>
<td>Laminar prem.</td>
<td>C₂H₄/air</td>
<td>1 bar</td>
<td>0D TiRe LII</td>
<td>Hot</td>
</tr>
<tr>
<td>Laminar prem.</td>
<td>C₂H₂/air</td>
<td>1 bar</td>
<td>1D + TiRe LII</td>
<td>Hot</td>
</tr>
<tr>
<td>Laminar diff.</td>
<td>C₂H₄</td>
<td>1 bar</td>
<td>0D TiRe 2C-LII</td>
<td>Hot</td>
</tr>
</tbody>
</table>
## Experiments performed

<table>
<thead>
<tr>
<th>Experiment Type</th>
<th>Substance &amp; Conditions</th>
<th>Pressure Range</th>
<th>Conditions</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laminar diffusion CH&lt;sub&gt;4&lt;/sub&gt;</td>
<td>1-80 bar</td>
<td></td>
<td>0D TiRe 2C-LII</td>
<td>hot</td>
</tr>
<tr>
<td>Quenched laminar diffusion C&lt;sub&gt;2&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;</td>
<td>1 bar</td>
<td></td>
<td>0D TiRe 2C-LII</td>
<td>moderate</td>
</tr>
<tr>
<td>Sampled from laminar diffusion C&lt;sub&gt;2&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt; flame, soot diluted in He, Ne, Ar, Kr, Xe, N&lt;sub&gt;2&lt;/sub&gt;, CO&lt;sub&gt;2&lt;/sub&gt;, SF&lt;sub&gt;6&lt;/sub&gt;</td>
<td>1 bar</td>
<td></td>
<td>0D TiRe 2C-LII</td>
<td>ambient</td>
</tr>
<tr>
<td>Turbulent diesel and similar (engine)</td>
<td>High</td>
<td></td>
<td>0D TiRe LII</td>
<td>hot</td>
</tr>
<tr>
<td>Turbulent diesel (engine)</td>
<td>up to 75 bar</td>
<td></td>
<td>0D TiRe LII</td>
<td>hot</td>
</tr>
<tr>
<td>Turbulent diesel (engine)</td>
<td>50-70 bar</td>
<td></td>
<td>0D TiRe LII/2D simultaneously</td>
<td>hot</td>
</tr>
<tr>
<td>Soot generator from C&lt;sub&gt;3&lt;/sub&gt;O&lt;sub&gt;2&lt;/sub&gt;</td>
<td>1 bar Ar</td>
<td></td>
<td></td>
<td>ambient-hot</td>
</tr>
<tr>
<td>After flame</td>
<td>1 bar</td>
<td></td>
<td>2D</td>
<td>moderate</td>
</tr>
<tr>
<td>Carbon black</td>
<td>&lt; 10&lt;sup&gt;-3&lt;/sup&gt; mbar</td>
<td></td>
<td>0D TiRe LII/2D</td>
<td>ambient</td>
</tr>
<tr>
<td>Carbon black</td>
<td>1–30 bar</td>
<td></td>
<td>0D TiRe LII/2D simultaneously</td>
<td>ambient</td>
</tr>
<tr>
<td>Fe</td>
<td>Fe nanoparticle reactor</td>
<td>0.5 bar</td>
<td>0D TiRe LII</td>
<td>349 K</td>
</tr>
</tbody>
</table>
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 10µm 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

<table>
<thead>
<tr>
<th>Excitation / nm</th>
<th>Detection / nm</th>
<th>Interferences</th>
</tr>
</thead>
<tbody>
<tr>
<td>1064</td>
<td>405 ± 5 / 650 ± 5 (2C)</td>
<td>none</td>
</tr>
<tr>
<td>1064</td>
<td>441,6 ± 5 / 650 ± 25 (2C)</td>
<td>none</td>
</tr>
<tr>
<td>1064</td>
<td>400 / 700 (2C)</td>
<td>PAH, C&lt;sub&gt;2&lt;/sub&gt;</td>
</tr>
<tr>
<td>1064 (+532 for PAH)</td>
<td>Broadband vis or RG630</td>
<td>none</td>
</tr>
<tr>
<td>532</td>
<td>PAH, C&lt;sub&gt;2&lt;/sub&gt;</td>
<td>none</td>
</tr>
<tr>
<td>1064</td>
<td>400-650 (resolved)</td>
<td>C&lt;sub&gt;2&lt;/sub&gt; (?)</td>
</tr>
<tr>
<td>1064</td>
<td>550 / 694 (2C)</td>
<td>none</td>
</tr>
<tr>
<td>1064</td>
<td>460 / 650</td>
<td>none</td>
</tr>
<tr>
<td>1064</td>
<td>Broadband vis</td>
<td>none</td>
</tr>
<tr>
<td>1064</td>
<td>500 ± 12.5 / 700 ± 12.5</td>
<td>none</td>
</tr>
<tr>
<td>1064, 532</td>
<td>397±19 or 445±30 / 782±6 (2C)</td>
<td>PAH?, C&lt;sub&gt;2&lt;/sub&gt;?</td>
</tr>
<tr>
<td>1064, 532</td>
<td>445±30 / 750±25 (2C)</td>
<td>PAH?, C&lt;sub&gt;2&lt;/sub&gt;?</td>
</tr>
<tr>
<td>532</td>
<td>405</td>
<td>PAH</td>
</tr>
<tr>
<td>532</td>
<td>450 ± 20 / 650 ± 12.5</td>
<td>No PAH/C&lt;sub&gt;2&lt;/sub&gt; expected?</td>
</tr>
<tr>
<td>532</td>
<td>410</td>
<td>Particle size effects?</td>
</tr>
<tr>
<td>532</td>
<td>LII from … ?</td>
<td>none</td>
</tr>
</tbody>
</table>
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

- photomultiplier
- convex lens
- filter 694 nm
- beam splitter
- convex lens
- particles

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

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B.F. Kock, C. Schulz, P. Roth
IVG, University Duisburg-Essen
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 → very long \( \tau \)
- 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:
A. Boiarciuc, F. Foucher,
C. Mounaïm-Rousselle
University of Orléans

$\text{calibration} = \frac{<\text{Intensity}>}{\text{Soot fraction}}$
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
- \( E(m) = 0.232 + 1.2546 \times 10^{-4} \times \lambda(nm) \) @ 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 \( \lambda \) (Uni Lille)
- Mass spectroscopic study of PAH adsorbed on soot particles (Uni Lille)
- Influence of laser fluence on particle morphology or aggregate structure (\( \rightarrow \) 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 (\( \rightarrow \) Sandia) - extension desired *
- Defined in-situ soot generation: C3O2 pyrolysis (H-free soot!) – availability to LII researchers?
- Accommodation coefficient from shock tube experiments (Uni Duisburg) – commonly valid?
- Accommodation coefficient from noble gas (+others) experiments (NRC)

* see below
Influence of laser fluence on soot-particle size and morphology

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IVG, University Duisburg-Essen
Influence of laser fluence on soot-particle size and morphology

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Fast LII - LII Signal Evolution: Influence of Temporal Response

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.

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

<table>
<thead>
<tr>
<th>Exposure time:</th>
<th>0.5ns</th>
<th>2.0ns</th>
</tr>
</thead>
<tbody>
<tr>
<td>Accommodation coefficients:</td>
<td>$\alpha=0.28$</td>
<td>$\alpha=0.32$</td>
</tr>
<tr>
<td></td>
<td>$\alpha_v=0.9$</td>
<td>$\alpha_v=0.4$</td>
</tr>
<tr>
<td></td>
<td>$n=0.4$</td>
<td>$n=0.46$</td>
</tr>
</tbody>
</table>

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