Ultrashort Pulse Laser Diagnostics for Chemically Reacting Flows

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Optica Laser Systems Technical Group Webinar
October 14th, 2022
Laser Diagnostics for Reacting Flows: Selected Projects

- Femtosecond Laser-Induced Fluorescence (fs-LIF) for chemical species imaging
- Laser-Induced Breakdown Spectroscopy (LIBS) metal particles, propellants & explosives
- Digital Inline Holography (DIH)
- Planar Laser-Induced Fluorescence / Particle Image Velocimetry (PLIF/PIV)
- Femtosecond Coherent Anti-Stokes Raman Scattering (fs-CARS) for temperature imaging
- Ultrahigh-rate (10–1,000 kHz) pulse-burst lasers diagnostics
- Imaging diagnostics for high-rate materials testing & hypersonics
Optical Diagnostics and Imaging Laboratory: Some Applications

- Physical properties (T, P, v, ρ, x-y-z, ...)
- Chemical properties (species, reactions,..)
- Particles (size, shape, vel./acc., momentum,..)
Optical Diagnostics and Imaging Laboratory: Facilities

New Diagnostics Development:
Femtosecond (fs) Laser Lab (JCAIN 416)

Applied Laser Diagnostics:
(Turbomachinery Laboratory, Test Cell 133)
Webinar Outline

Ultrashort Pulse Laser Diagnostics for Chemically Reacting Flows

I. Laser Diagnostics 101 (*for gas-phase chemically reacting flows*)

II. Femtosecond Two-Photon Laser-Induced Fluorescence (Fs-TPLIF) of Atoms: *Basics & Some Applications*

III. Molecular Species Imaging: *CO & OH*

IV. Simultaneous Multi-Species Imaging Using a Single Femtosecond Laser (*H/OH, NO/O/O₂*)

*Ultrashort Pulse: Femtosecond-Duration (1 fs = 10⁻¹⁵ s)*
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The Electromagnetic Spectrum

Why ultrashort pulsed lasers?
Laser Sources for Spectroscopy/Optical Diagnostics
Laser Sources for Spectroscopy/Optical Diagnostics

Light Emitting Diode

![Light Emitting Diode Image](Image)

25 nm

BANDWIDTH

Wavelength (nm)

nm  500  700  900
Laser Sources for Spectroscopy/Optical Diagnostics

He-Ne cw laser

![He-Ne cw laser diagram](image)

Bandwidth

![Bandwidth graph](image)

Wavelength (nm)

nm

500  700  900

0.002 nm
Laser Sources for Spectroscopy/Optical Diagnostics

Nd:YAG Laser (ns)

Pulsed!
(Typically, 10 Hz)
Laser Sources for Spectroscopy/Optical Diagnostics

Ti:Sapphire modelocked oscillator/amplifier (fs)

Pulsed! (1-10 kHz)

Heisenberg Uncertainty Principle

\[ \Delta \nu \times \Delta t = \text{Const (0.315)} \]
Ultrashort Pulse (Femtosecond) Laser Sources for Spectroscopy/Optical Diagnostics

Femtosecond (fs)-duration pulses \(1 \text{ fs} = 10^{-15} \text{ s}\)

Optical Parametric Amplifiers

Amplified Ti:Sapphire Laser systems (35, 80, 100 fs)

Harmonic Generation (home-built)
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Atomic species play critical roles in combustion/plasma systems due to high reactivity/diffusivity

Thermal NO₂ Formation: Zeldovich Mechanism

\[ \text{N}_2 + \text{O} \rightarrow \text{NO} + \text{N} \]
\[ \text{N}_2 + \text{O}_2 \rightarrow \text{NO} + \text{O} \]
\[ \text{N} + \text{OH} \rightarrow \text{NO} + \text{H} \]


Soot Formation in Hydrocarbon Flames

“H-Abstraction-C₂H₂-Addition” (HACA)

\[ \text{n-C}_2\text{H}_4 + \text{C}_2\text{H}_2 \rightarrow \text{benzene} + \text{H} \]
\[ \text{C}_2\text{H}_4 + \text{C}_2\text{H}_2 \rightarrow \text{benzene or phenyl} + \text{H} \]
\[ \text{C}_2\text{H}_4 + \text{CH}_2 \rightarrow \text{benzene} + \text{H} + \text{H} \]
\[ \text{A}_1 + \text{H} \rightarrow \text{A}_1 + \text{H}_2 \]
\[ \text{A}_1 + \text{C}_2\text{H}_2 \rightarrow \text{A}_1\text{C}_2\text{H}_2 \]
\[ \text{A}_1\text{C}_2\text{H}_2 + \text{C}_2\text{H}_2 \rightarrow \text{A}_1\text{C}_2\text{H}_4 + \text{H} \]

A₁: Aromatic compound


Plasma-Assisted Hydrocarbon Fuel Reforming (i.e. JP-8)

\[ \text{C}_n\text{H}_m + e^- \rightarrow \text{C}_n\text{H}_{m-1} + \text{H} + e^- \]
\[ \text{H} + \text{O}_2 \rightarrow \text{OH} + \text{O} \]

Cappelli et al., Combust. Flame. 153, 603 (2008)

Rotating Non-Equilibrium Gliding Arc Plasma Disc for Enhancement in Ignition and Combustion


Bacterial Inactivation

Plasma Medicine
Single-photon transitions lie in VUV; Multi-photon excitation schemes are required

<table>
<thead>
<tr>
<th>Species</th>
<th>Excitation Wavelength (nm)</th>
<th>LIF Wavelength (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>2 x 205</td>
<td>656</td>
</tr>
<tr>
<td>O</td>
<td>2 x 226</td>
<td>845</td>
</tr>
<tr>
<td>N</td>
<td>2 x 207</td>
<td>745</td>
</tr>
<tr>
<td>Kr</td>
<td>2 x 204</td>
<td>826</td>
</tr>
<tr>
<td>Xe</td>
<td>2 x 224</td>
<td>835</td>
</tr>
<tr>
<td>CO</td>
<td>2 x 230</td>
<td>483</td>
</tr>
</tbody>
</table>

- Collisional quenching
- Photoionization
- Stimulated emission
- Photolytic production

During TPLIF, intense laser pulses can photochemically produce atomic hydrogen.

- Main photolytic precursors for H:
  - H₂O (H₂O +hv -> H + OH)
  - CH₃ (CH₃ +hv -> CH₂ + H)
    - CH₂ +hv -> CH + H
    - CH +hv -> C + H)

H-atom LIF line profile (ns excitation)

Axi-symmetric, premixed, CH₄/O₂/N₂ flame

Interference-free line imaging of H atoms using picosecond and femtosecond Pulses

Premixed, CH$_4$/O$_2$/N$_2$ flame

**ns excitation** (~8 ns)

**ps excitation** (~100 ps)


10 kHz, Single-shot


10-20 Hz, Shot-averaged
Femtosecond Two-Photon LIF (fs-TPLIF)

- Broad bandwidth of fs pulses contribute to efficient TP excitation
- High peak power is ideal for multi-photon excitation
- Reduced photolytic interferences
- kHz-rate multi-dimensional imaging capability
- Extension to collisional-quenching-free detection

Efficient two-photon excitation

Femtosecond CARS


Typical Experimental Apparatus

**Laser System**
- Ultrafast amplifier (80 fs FWHM, 7 mJ/pulse at 1 kHz)
- Femtosecond OPA (~8 μJ/pulse at 205 nm)

**Imaging System**
- High-speed visible intensifier, lens coupled to a CMOS camera (or CCD) for kHz-rate imaging
- Also use an intensified CCD (ICCD) camera (~30 Hz)
Fs-TPLIF of Atomic Species in Reacting Flow/Fluid Dynamic Studies

TPLIF Excitation Schemes:

<table>
<thead>
<tr>
<th>Element</th>
<th>Excitation Scheme</th>
<th>Wavelengths</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>3d $^2D_{3/2,5/2}$, 656.5 nm</td>
<td>3p $^3P_{1,2,0}$, 844.9 nm</td>
</tr>
<tr>
<td>O</td>
<td>2p $^4P_{1/2,3/2}$, 2×205.1 nm</td>
<td>3s $^3S$, 742-747 nm</td>
</tr>
<tr>
<td>N</td>
<td>1s $^2S_{1/2}$, 2×206.7 nm</td>
<td>2p $^3S_{1/2}$, 2×204.2 nm</td>
</tr>
<tr>
<td>Kr</td>
<td>5p$^6[3/2]_2$, 826.5 nm</td>
<td>6p$^6[3/2]_2$, 834.9 nm</td>
</tr>
<tr>
<td>Xe</td>
<td>6s$^6[1/2]_1$, 2×224.3 nm</td>
<td></td>
</tr>
</tbody>
</table>

Fs- TPLIF Applications (selected)

- **Cellular Flames (H-atom)**

- **Atmos. P Plasma Jets (O-atom)**

- **Jet Flame (O-atom SS)**

- **Flow Mixing (Kr PLIF)**

Simultaneous Multiple Species Imaging

- **Simultaneous H/OH Imaging**
Experimental Apparatus for High-Pressure H-Atom Imaging

Typical frequency conversion efficiencies:
- Commercial OPAs ~ 0.1 to 0.2%
- FHG ~ 2.5% (*home-built*)

Laser Transmission Losses Through Thick Optical Windows

- Transmission efficiency decreased from ~80-20% as the energy increased from 1–50 μJ/pulse.
- Nonlinear effects at the quartz window and focusing lens.
- Losses overcome by the high conversion efficiency of home-built FHG.

Laser Energy Dependence

- Focusing lens used, \( f = +200\)-mm, estimated beam waist \( \sim 100 \, \mu\text{m} \).
- No apparent deviation from quadratic dependence.
- Negligible photoionization and stimulated emission (SE) in flames investigated.

Amplified Spontaneous Emission (ASE) Study

- Sharp onset of ASE as the pulse energy increased
- Pulse stretching by the window material appears to reduce ASE
- ASE interference-free measurements for all pressures during this study

H-Atom LIF Radial Profiles

UNICORN model results courtesy of Dr. Vish Katta (ISSI)


Hencken Burner CH₄/Air Flame (Φ = 1.2)

- Chemical reaction takes place in the inner premixed region
- Radial H profiles show good agreement with the model predictions
- Slight discrepancy at P=10 bar attributed to buoyancy-driven flame flickering
Two-Dimensional Distribution of H-Atom

- Single TPLIF planar images of H not feasible at high-pressure due to low laser energy
- H-atom line images recorded by translating the burner vertically in 0.35-mm steps
- Good match between experiment and numerical predictions

Example Case:
P = 2 bar

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Molecular Species Imaging: Carbon Monoxide (CO) TPLIF

CO TPLIF Excitation/Detection

CO fs-TPLIF: Experimental

Pure CO in a gas cell at 1 bar

Strong pressure and quenching


CO TPLIF spectrum in flames: C₂ interferences

C$_2$ Interferences: Detection Gate Width & Spectral Filtering

- C$_2$ cross-talk reduces available CO detection lines
- Narrower detection gate width (2–10 ns) reduces C$_2$ interference and as well as LIF signal

Premixed C$_2$H$_4$-air flame with equivalence ratio $\Phi=1.5$

Fs-TPLIF of CO in Methane and Ethylene (Sooting) Flames

Premixed CH$_4$/Air Flame

Flames operated on near-adiabatic Hencken calibration burner at 1 atm

Fs-TPLIF of CO in Sooting $C_2H_4/O_2/N_2$ Flames

Single-Shot Images

Beam sheet height ~5 mm

CO Imaging in Piloted Liquid-Fuel Spray Flames

Multi-Fuel Operation

Modified McKenna Burner
Pilot Flame: CH₄/Air

Two-Color OH
PLIF Temp

Molecular Species Imaging: Hydroxyl Radical (OH) PLIF (single-photon excitation)

**Experimental Apparatus**

**OH Excitation Spectrum**

**1-kHz OH PLIF in Turbulent Flames**

**High-Resolution Emission Spectrum**

Wang, Kulatiaka et. al., Comb. Flame, 214 (2020)
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Excitation and Detection Scheme for NO, O, & O₂

• NO excitation/detection
  • NO A-X (0,0) excited at 226 nm. Reduce O₂ interferences at this wavelength
  • Strong emission at 236- and 247-nm bands

• O excitation
  • 3p³P ↔ 2p³P two-photon transition at 226 nm
  • 845-nm emission from 3p³P → 3s³S decay

• O₂ excitation
  • B³–X³Σ⁻g Schumann-Runge system
  • Highly temperature dependent
  • Excited by laser radiation from 175–535 nm
  • Various emission lines from 200–400 nm

Experimental Apparatus for Simultaneous NO, O, & O₂ Detection

- **Laser System**
  - Ti:Sapphire Laser 800 nm ~ 80 fs pulse duration
  - TOPAS Prime OPA generating ~226 nm pulses with maximum energy of 14 μJ/pulse, FWHM ~2.3 nm
  - Three mirrors and a f = +500 mm lens (L1) focused the beam at the center of the flame

- **Imaging Systems**
  - Spectrometer and UV ICCD Camera
  - Spectral Mode: Capture emission spectra of all three species
  - Imaging Mode: Captured NO and O₂ line images
  - Direct Imaging with visible ICCD Camera for single-shot O- atom detection

NO, O, & O₂ Excitation and Detection Scans

- Laser excitation wavelength scan:
  - OPA output wavelength scanned from 220–228.5 nm
  - 226.1 nm radiation maximizes NO and O-atom signal. Still produces some O₂ excitation
  - NO and O₂ experimental data compared to convolutions of LIFSIM simulations

- Laser energy scan:
  - Excitation wavelength fixed at 226.1 nm
  - Linear energy dependence on laser energy for NO and O₂
  - Quadratic dependence on laser energy for O-atom

![Diagram showing laser excitation and detection scans](image-url)
NO, O, & O₂ Detection Spectral Characterization

- NO, O, and O₂ LIF signals collected through an imaging spectrometer (300 lines/mm and 1200 lines/mm gratings)

- Interference-free emission lines for NO and O₂:
  - NO: 236, 247, and 285 nm
  - O₂: 241, 250, 278, and 289 nm

- All scans conducted with 226.1-nm excitation and 14-µJ/pulse laser energy

- Estimated NO detection limit between 10~40 ppm in flames

NO, O, & O₂ Detection Spectral Characterization – 1D Imaging

- NO and O₂ imaged through the spectrometer. Emission wavelength (X-axis) radial distance (Y-axis)

- Spectra above each image created by full-vertical binning

- NO increases with increasing height-above-burner (HAB). O₂ decreases as it is consumed by the flame

NO, O, & O₂ Measurements in a Range of Flame Conditions

- Flame equivalence ratio (Φ) scans conducted. Data recorded at the center of the flame at 20-mm HAB
- Numerical predictions performed using Cantera simulation code with GRI 3.0 chemical mechanism
- Quenching correction considering CO, N₂, H₂, CH₄, H₂O, CO₂, and O₂ as colliders
- Departure in rich NO measurements is likely due to prompt NO formation

In Summary,

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II. Fs-TPLIF of Atomic Species: Basics & Some Applications

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Acknowledgements

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- Morgan O’Neil
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Q & A