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OPTICA | Formerly OSA

Ultrashort Pulse Laser Diagnostics for Chemically Reacting Flows

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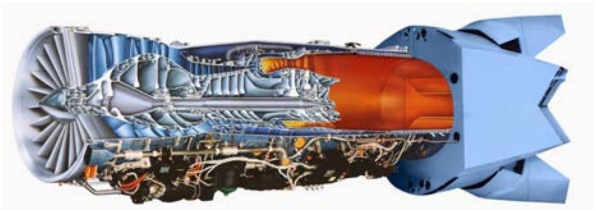
College Station, TX 77843, U.S.A.

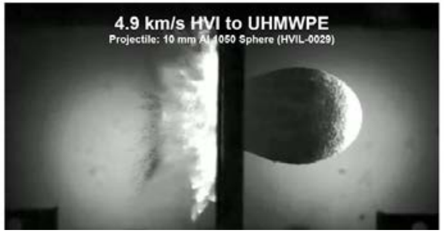
Optica Laser Systems Technical Group Webinar
October 14^{th}, 2022

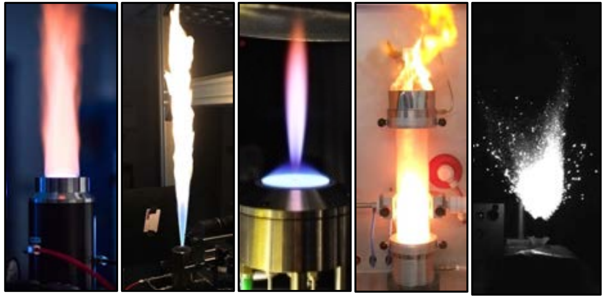
Laser Diagnostics for Reacting Flows: Selected Projects

- 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







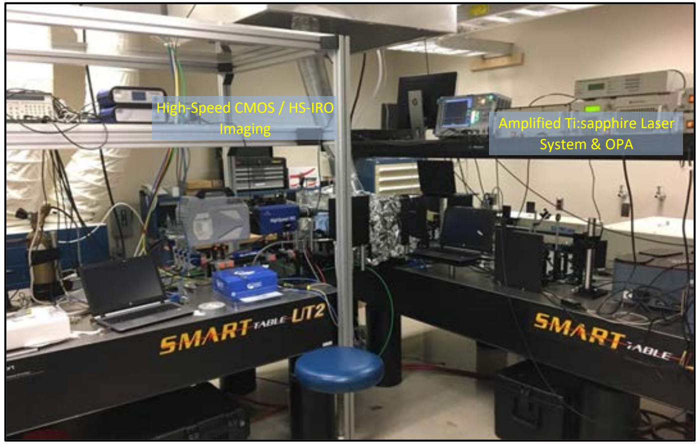
Optical and laser-based measurement

- 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

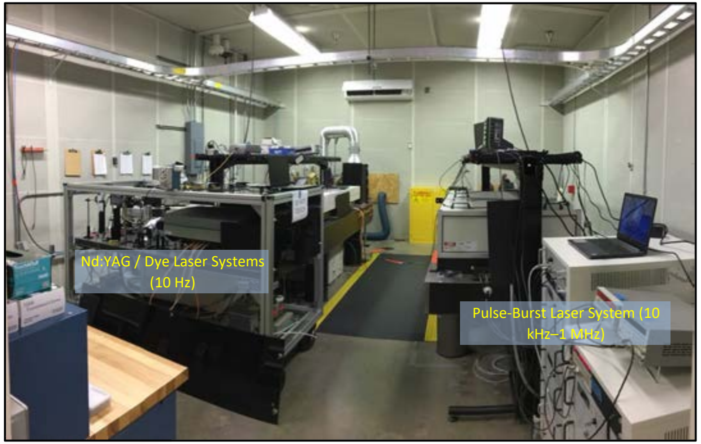
New Diagnostics Development:

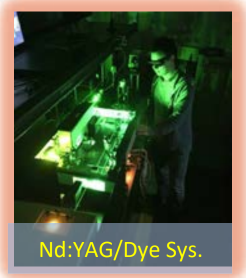
Femtosecond (fs) Laser Lab (JCAIN 416)



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/O_2)*

**Ultrashort Pulse: Femtosecond-Duration (1 fs = 10^{-15} s)*

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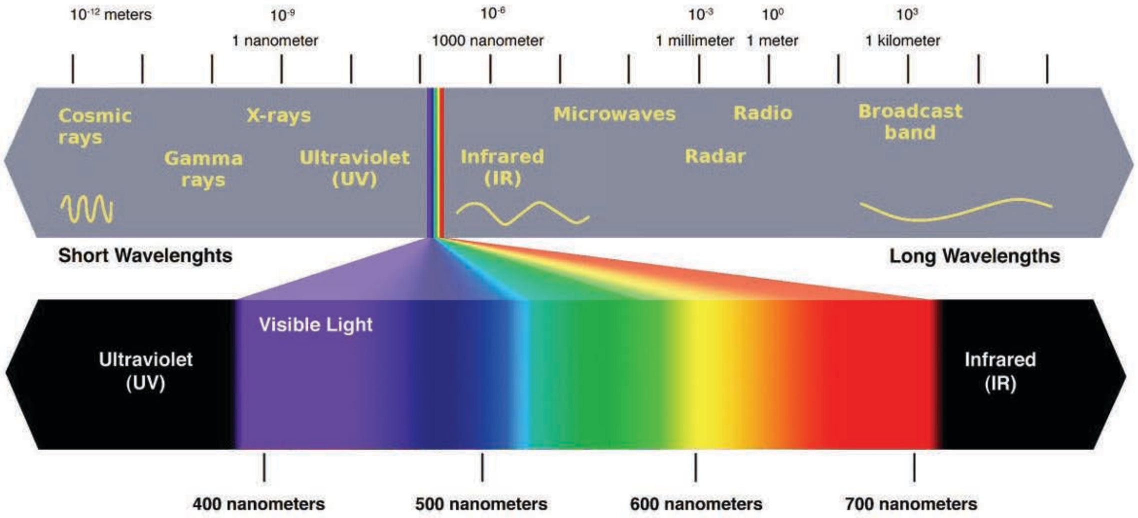
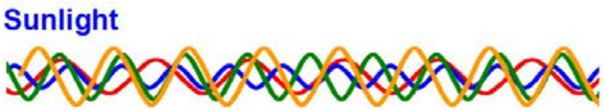
Ultrashort Pulse: Femtosecond-Duration (1 fs = 10^{-15} s)

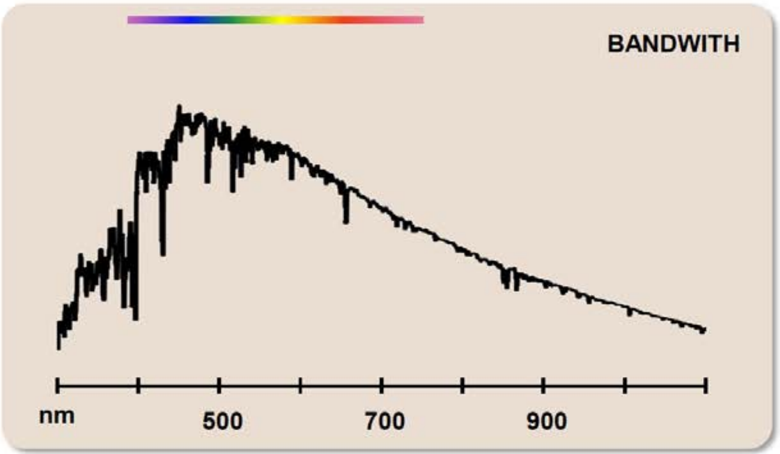
The Electromagnetic Spectrum



Why ultrashort pulsed lasers?

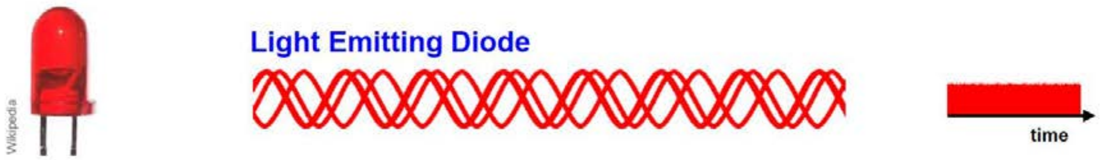
Laser Sources for Spectroscopy/Optical Diagnostics

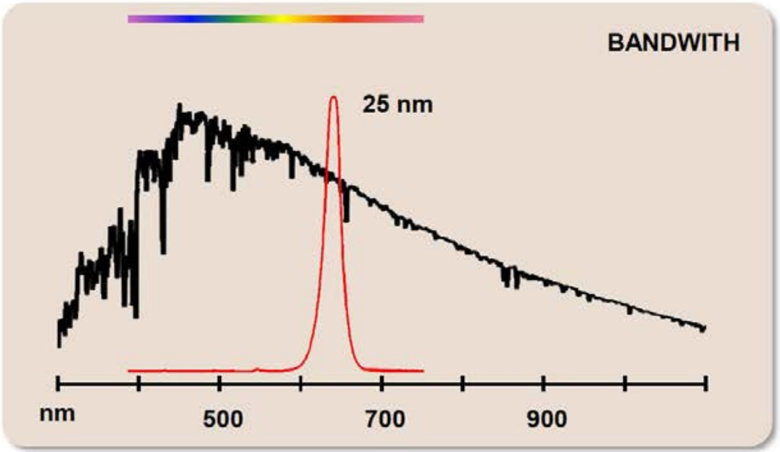




Wavelength (nm)

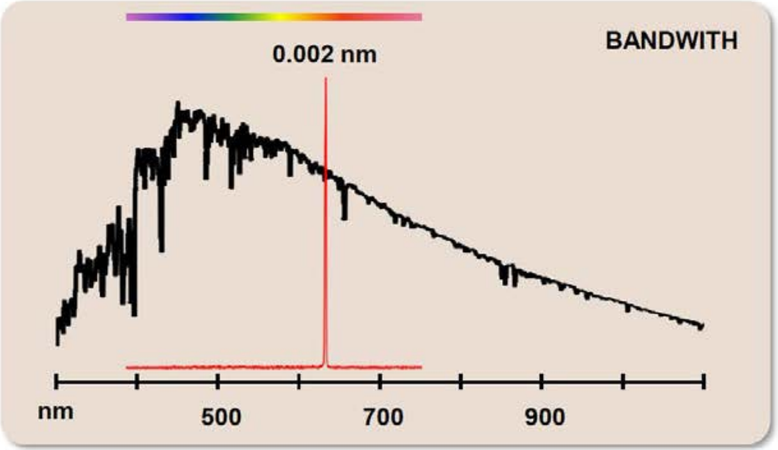
Laser Sources for Spectroscopy/Optical Diagnostics





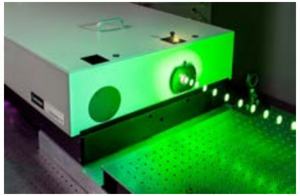
Wavelength (nm)

Laser Sources for Spectroscopy/Optical Diagnostics

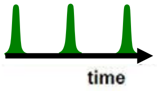


Wavelength (nm)

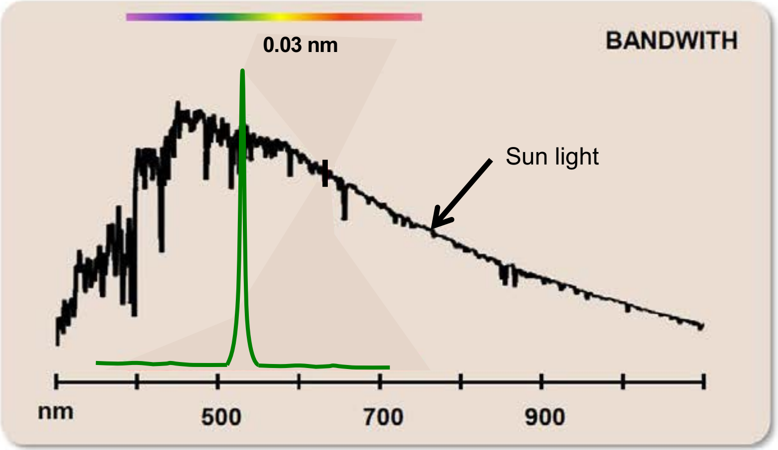
Laser Sources for Spectroscopy/Optical Diagnostics



Nd:YAG Laser (ns)



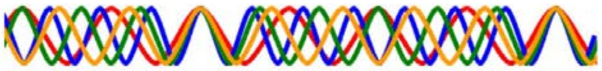
Pulsed !
(Typically, 10 Hz)

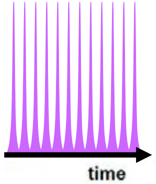


Wavelength (nm)

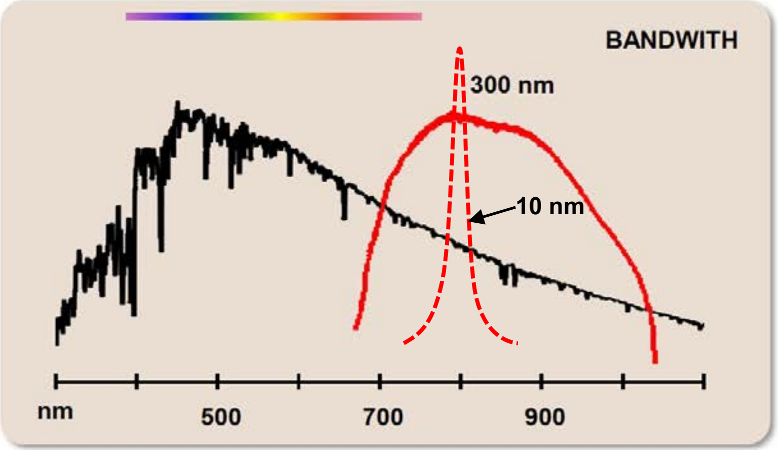
Laser Sources for Spectroscopy/Optical Diagnostics

Ti:Sappire modelocked oscillator/amplifier (fs)

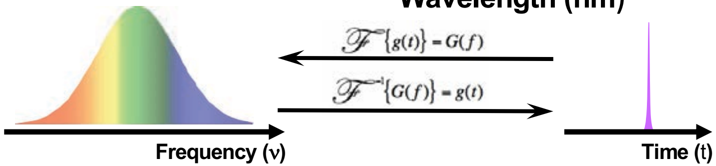




Pulsed !
(1-10 kHz)



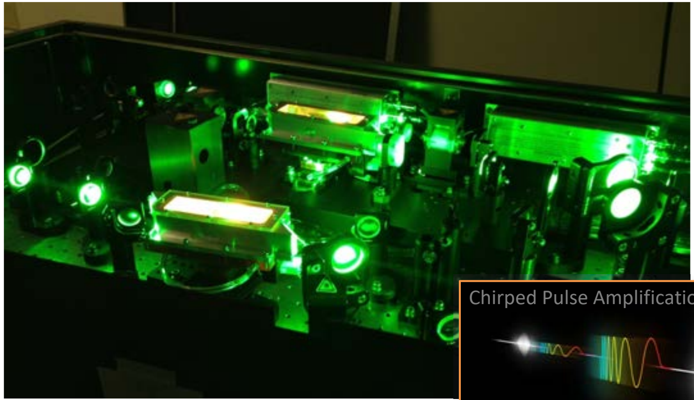
Wavelength (nm)

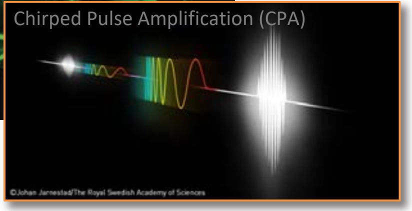


Heisenberg Uncertainty Principle

$\Delta\nu * \Delta t = \text{Const } (0.315)$

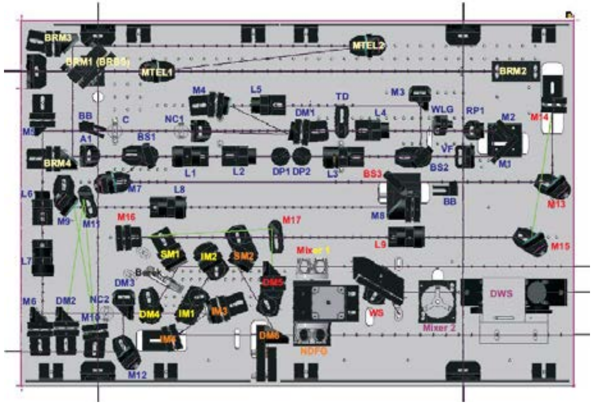
Ultrashort Pulse (Femtosecond) Laser Sources for Spectroscopy/ Optical Diagnostics





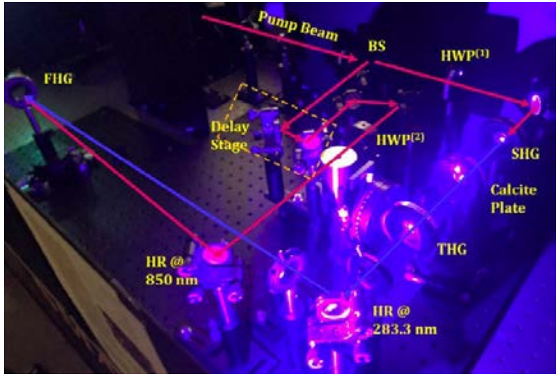
Femtosecond (fs)-duration pulses
{1 fs = 10^{-15} s}

Optical Parametric Amplifiers



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

Harmonic Generation *(home-built)*



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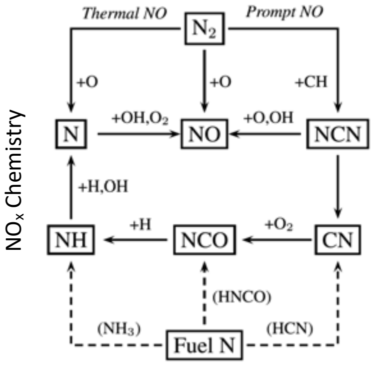
**Ultrashort Pulse: Femtosecond-Duration (1 fs = 10^{-15} s)*

Atomic species play critical roles in combustion/plasma systems due to high reactivity/diffusivity



Thermal NO_x Formation:
Zeldovich Mechanism

$N_2 + O \rightarrow NO + N$

$N + O_2 \rightarrow NO + O$

$N + OH \rightarrow NO + H$

Glaborg, *Proc. Combust. Inst.* **31**, 77 (2006)

Soot Formation in Hydrocarbon Flames

"H-Abstraction-C_2H_2-Addition" (HACA)

$n\text{-}C_4H_5 + C_2H_2 \rightarrow \text{benzene} + H$ (1)

$C_3H_3 + C_3H_3 \rightarrow \text{benzene} \quad \text{or} \quad \text{phenyl} + H$ (2)

$C_5H_5 + CH_3 \rightarrow \text{benzene} + H + H$ (3)

$A_i + H \rightleftharpoons A_{i-} + H_2$ (4)

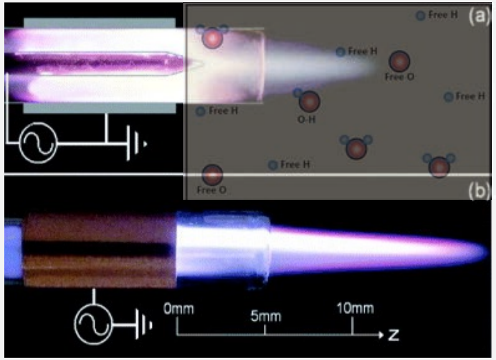
$A_{i-} + C_2H_2 \rightleftharpoons A_iC_2H_2$

$A_iC_2H_2 + C_2H_2 \rightarrow A_{i+1} + H$ (5)

A_i : Aromatic compound

H. Wang and K. Brezinsky, J. Phys. Chem., 1998, 102, 1530

Plasma Medicine



Bacterial Inactivation

Kong et al. *New J. Phys.* **11**, 115012 (2009)

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

$C_nH_m + e^- \rightarrow C_nH_{m-1} + H + e^-$

$H + O_2 \rightleftharpoons OH + O$

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

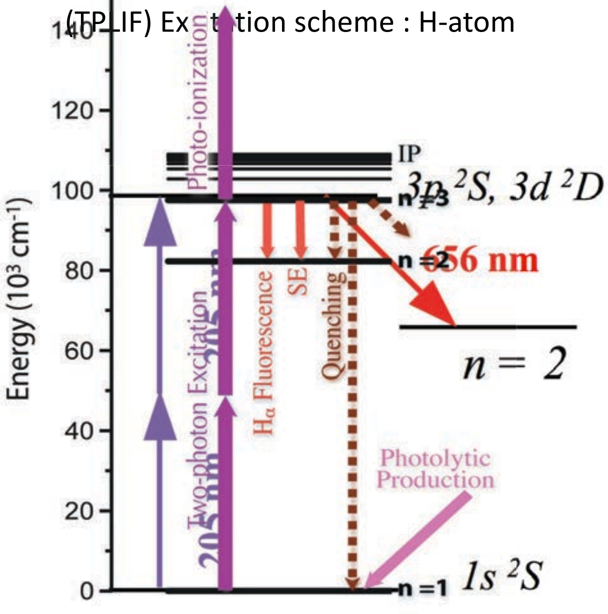
Rotating Non-Equilibrium Gliding Arc Plasma Disc for Enhancement in Ignition and Combustion

Gallagher M., *PhD Thesis – Drexel Univ.* (2010)

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

Two-photon laser-induced fluorescence (TPLIF) Excitation scheme : H-atom

H-atom TPLIF



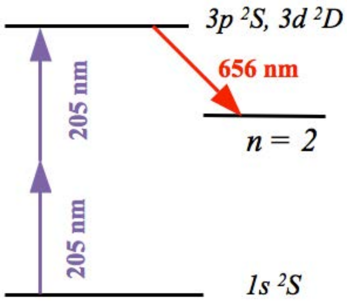
Lucht, Larendeau et al. *Opt. Lett.* **8,** 365-
Goldsmith *Opt. Lett.* **11,** 416-418 (1986)
Bittner, Kohse-Höinghaus et al. *Combust. Flame* **71,** 41-50 (1988)
Döbele et al., *Plasma Sources Sci. Technol.* **14**, S31–S41 (2005)
Frank & Settersten, Proc. Combust. Inst., **30,** 1527–1534 (2005)
Brackmann, Aldén et al., *Proc. Combust Inst.* **34**, 3541–3548 (2013)

Other species requiring TP excitation

Species	Excitation Wavelength (nm)	LIF Wavelength (nm)
H	2 x 205	656
O	2 x 226	845
N	2 x 207	745
Kr	2 x 204	826
Xe	2 x 224	835
CO	2 x 230	483

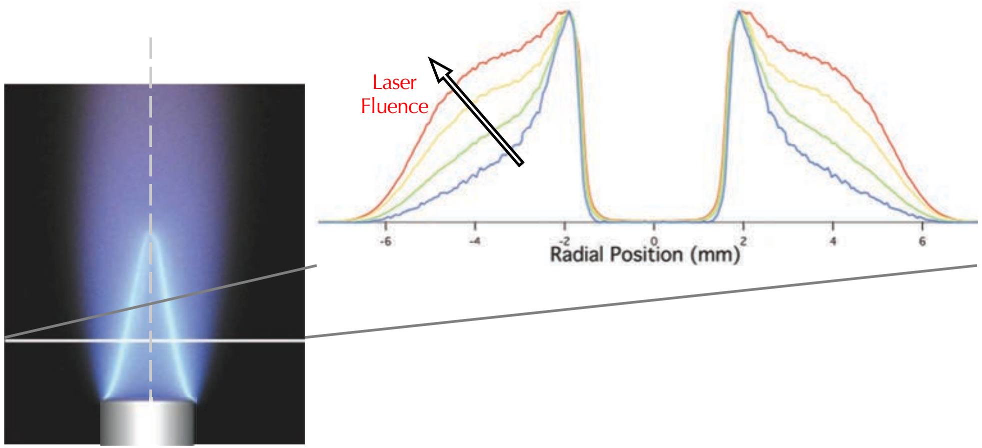
- Collisional quenching
- Photoionization
- Stimulated emission
- Photolytic production

During TPLIF, intense laser pulses can photochemically produce atomic hydrogen



H atom

H-atom LIF line profile (ns excitation)



Axi-symmetric, premixed, $CH_4/O_2/N_2$ flame

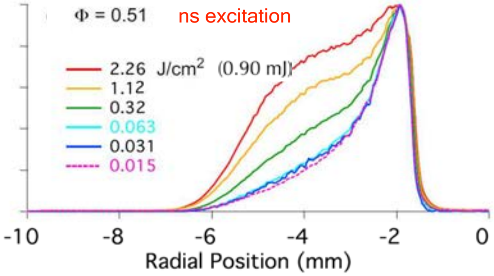
- Main photolytic precursors for H:

  - H_2O (H_2O + hν -> H + OH)
  - CH_3 (CH_3 + hν -> CH_2 + H
    - CH_2 + hν -> CH + H
    - CH + hν -> C + H)

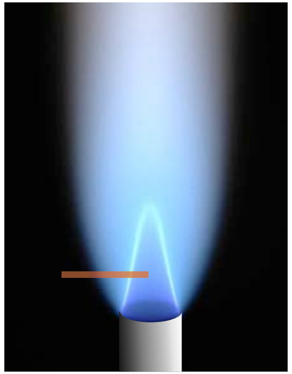
Goldsmith *Opt. Lett.* **11,** 416-418 (1986)
Frank, Settersten et. al., *App. Opt.*, **43,** 2588–2597 (2004).
Kulatilaka, Settersten, Frank et al., *Appl. Phys. B.* ***97***, 227-242 (2009)

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

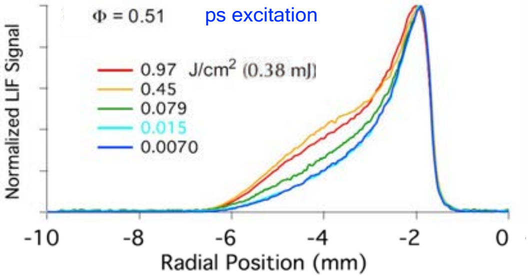
ns excitation (~8 ns)



Premixed, $CH_4/O_2/N_2$ flame

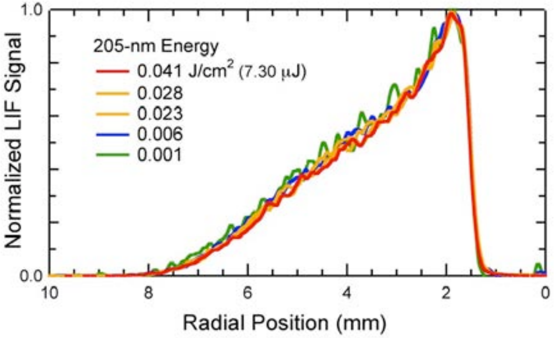


ps excitation (~100 ps)



Kulatilaka, Settersten, Frank et al., *Appl. Opt.* **47,** 4672-4683 (2008)

fs excitation (~100 fs)

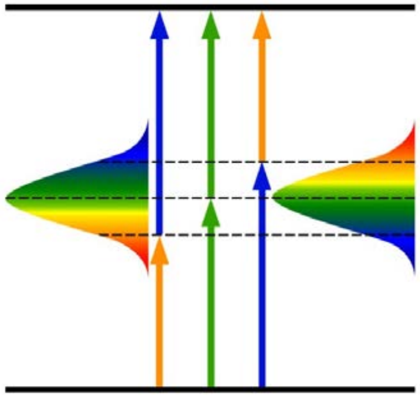


Kulatilaka, et. al., Opt. Lett., **37,** 3051-3053 (2012)

Femtosecond Two-Photon LIF (fs-TPLIF)

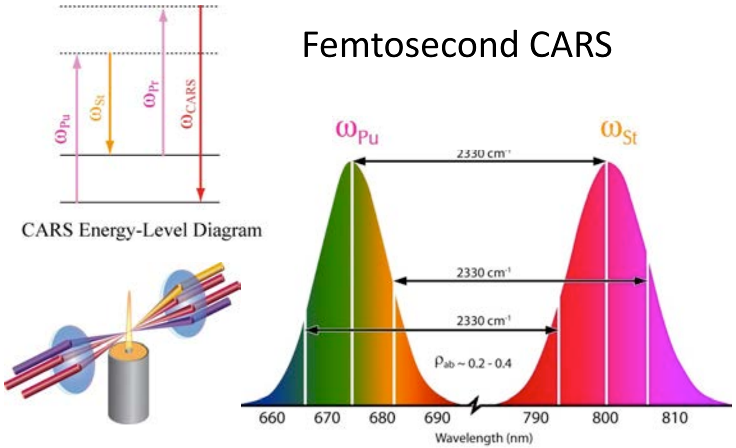
- Broad bandwidth of fs pulses contribute to efficient TP excitation
- High peak power is ideal for multi-photon excitation
- Reduced photolytic interferences
- kHz-rate multi-dimensional imaging capability
- Extension to collisional-quenching-free detection

Efficient two-photon excitation



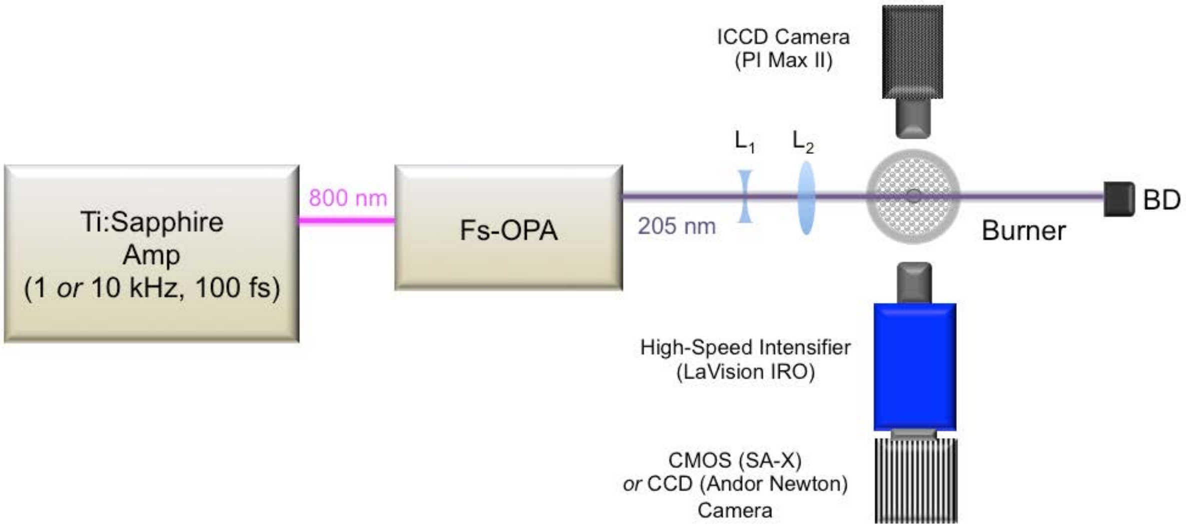
Kulatilaka, et. al., Opt. Lett., **37,** 3051-3053 (2012)

Femtosecond CARS



- Motzkus et al. *J Chem . Phys.,* **115,** 5418-5426 (2001)
- Roy, Kulatilaka et al., *Opt. Lett.,* **34,** 3857-3859 (2011)
- Kearney & Danehy, *Opt. Lett,* **40**, 4082–4085 (2015)
- Bohling & Kliewer, *J. Chem. Phys.,* **138,** 221101 (2013)

Typical Experimental Apparatus



Laser System

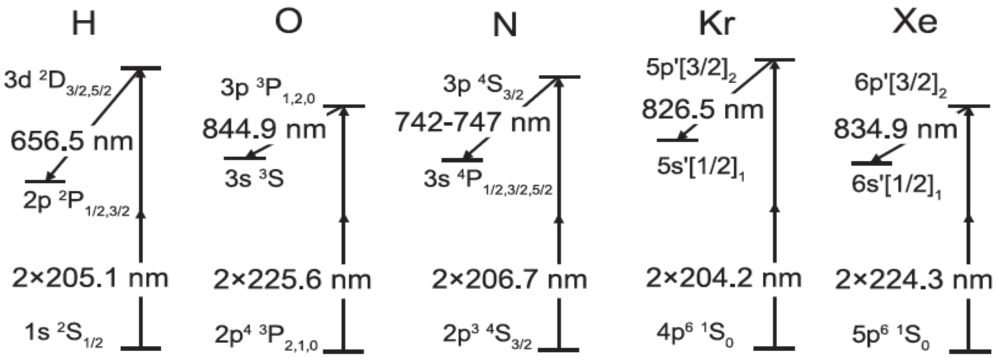
- Ultrafast amplifier (80 fs FWHM, 7 mJ/pulse at 1 kHz)
- Femtosecond OPA (~8 μJ/pulse at 205 nm)

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)

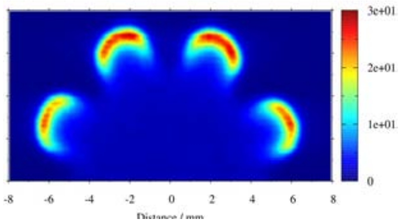
Fs-TPLIF of Atomic Species in Reacting Flow/Fluid Dynamic Studies

TPLIF Excitation Schemes:

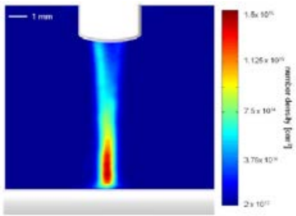


Fs- TPLIF Applications (selected)

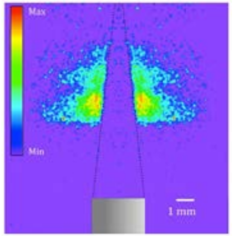
Simultaneous Multiple Species Imaging



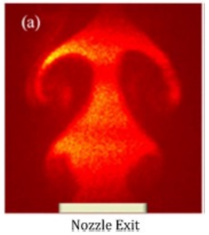
Cellular Flames (H-atom)
Hall, Kulatilaka, Pitz, Gord, *Proc. Combust. Inst.*, **35,** (2015)



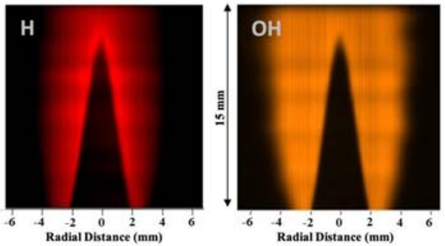
Atmos. P Plasma Jets (O-atom)
Schmidt, Kulatilaka, Gord et al., *PSST,* **24,** (2015)



Jet Flame (O-atom SS)
Kulatilaka, Gord, et al., *Appl. Phys. B,* **122,** (2016)

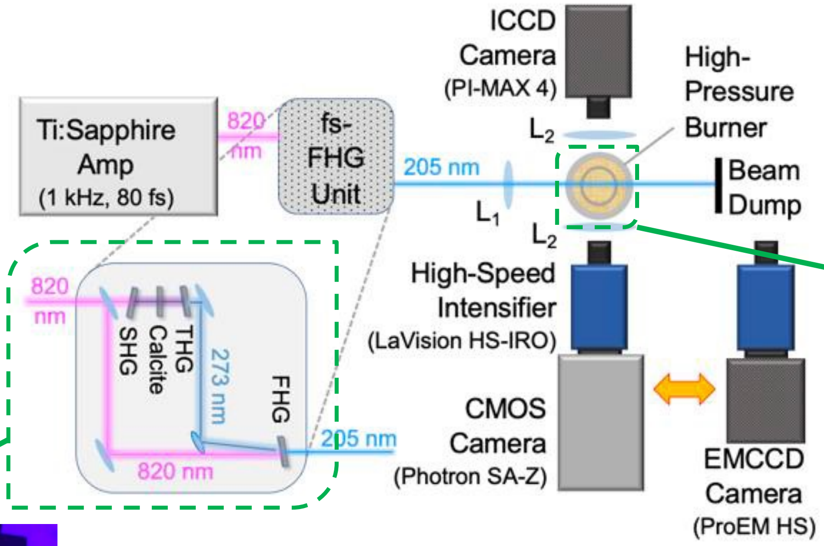


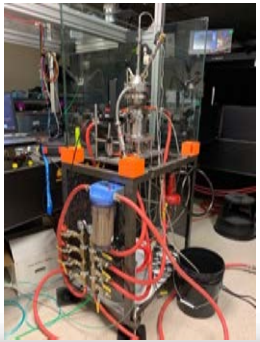
Flow Mixing (Kr PLIF)
Wang, Kulatilaka, *Opt. Lett.,* **42,** (2017)

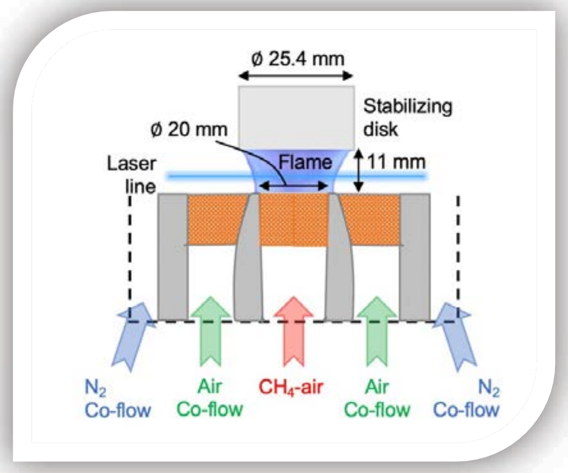


Simultaneous H/OH Imaging
Jain, Wang, Kulatilaka, *Proc. Combust. Inst (2021)*
Parajuli, Jain, Wang, Kulatilaka, *ASME Turbo Expo (2020)*

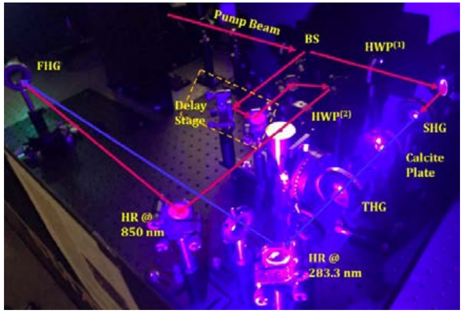
Experimental Apparatus for High-Pressure H-Atom Imaging





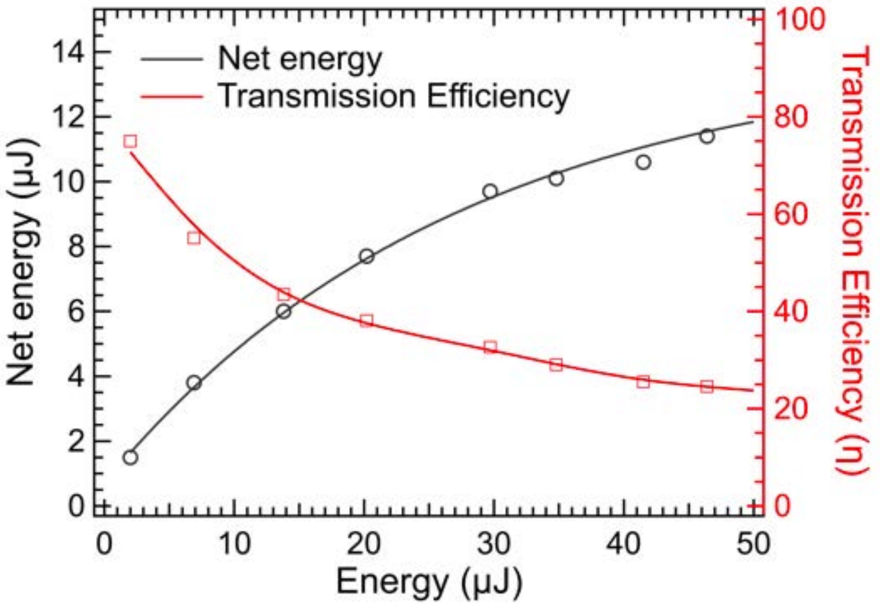


Parajuli, Kulatilaka, et al., *Proc. Combust. Inst.* In Press. (2023)



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

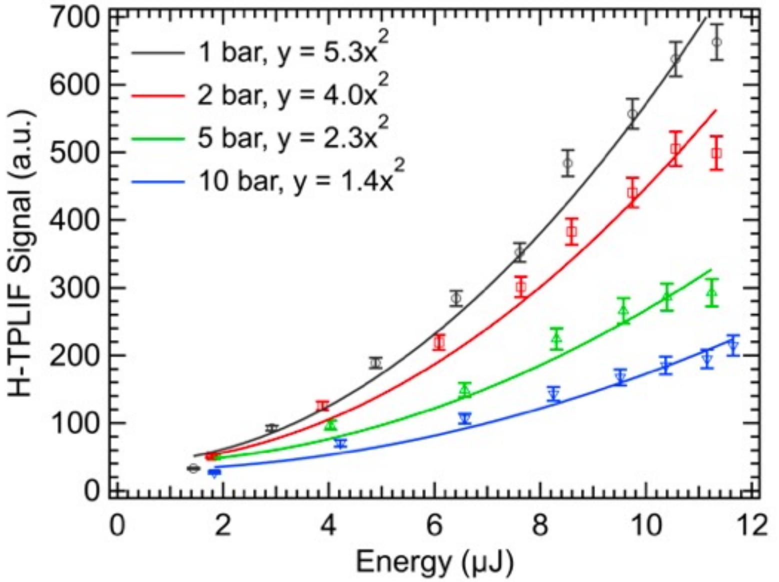
Laser Transmission Losses Through Thick Optical Windows



Parajuli, Kulatilaka, et al., *Proc. Combust. Inst.* In Press. (2023)

- 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

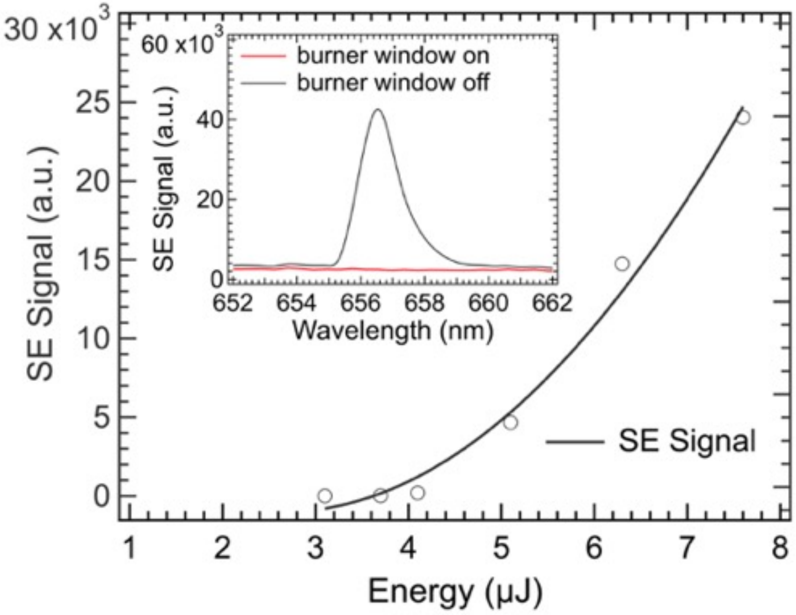
Laser Energy Dependence



Parajuli, Kulatilaka, et al., *Proc. Combust. Inst.* In Press. (2023)

- Focusing lens used, f= +200-mm, estimated beam waist ~ 100 μm.
- No apparent deviation from quadratic dependence.
- Negligible photoionization and stimulated emission (SE) in flames investigated

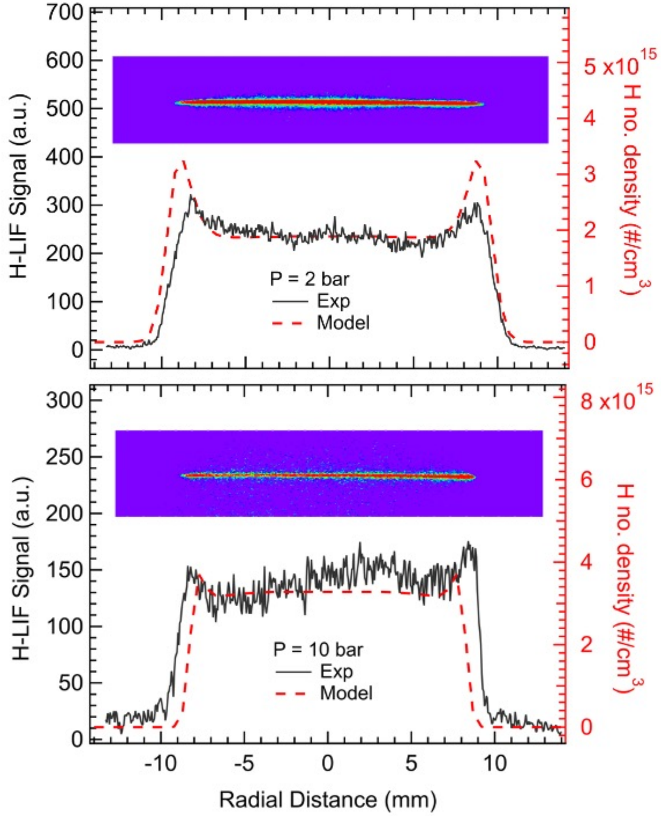
Amplified Spontaneous Emission (ASE) Study

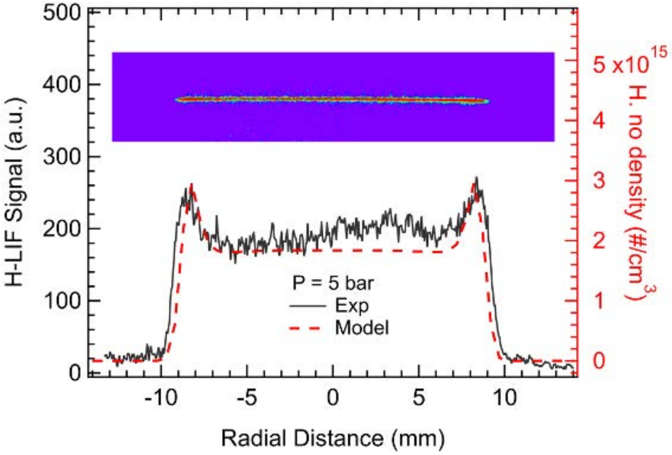


Parajuli, Kulatilaka, et al., *Proc. Combust. Inst.* In Press. (2023)

- 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)

Parajuli, Kulatilaka, et al., *Proc. Combust. Inst.* In Press. (2023)

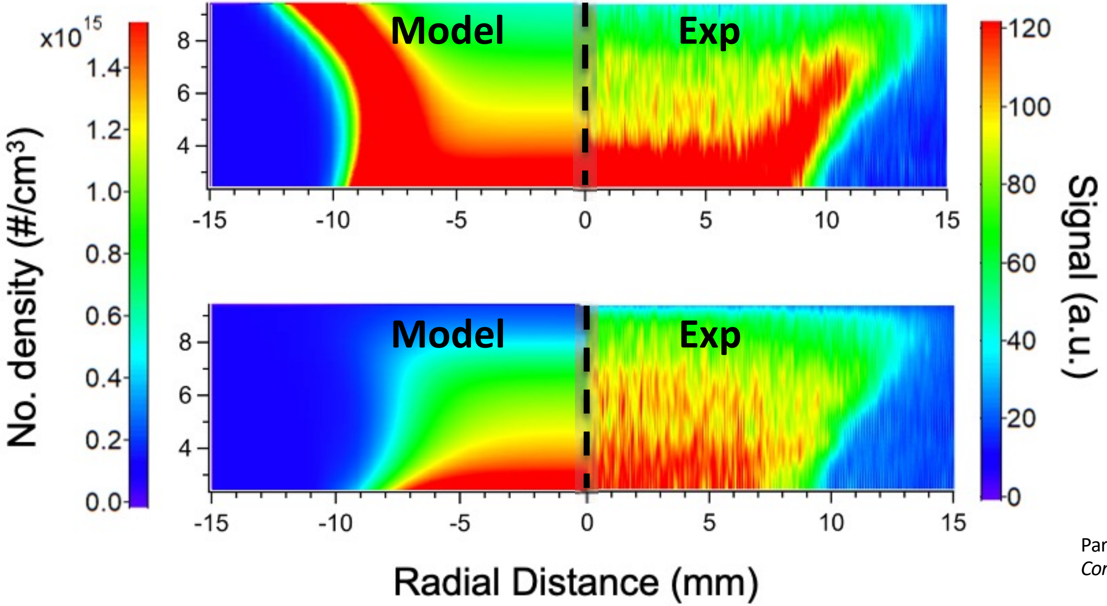
Hencken Burner CH_4/Air Flame ($\Phi = 1.2$)

- 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



Parajuli, Kulatilaka, et al., *Proc. Combust. Inst.* In Press. (2023)

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*

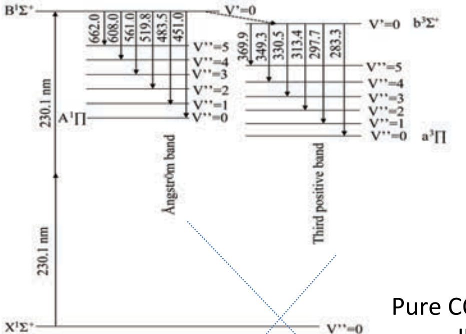
III. Molecular Species Imaging: *CO & OH*

IV. Simultaneous Multi-Species Imaging Using a Single Femtosecond Laser *(H/OH, NO/O/O_2)*

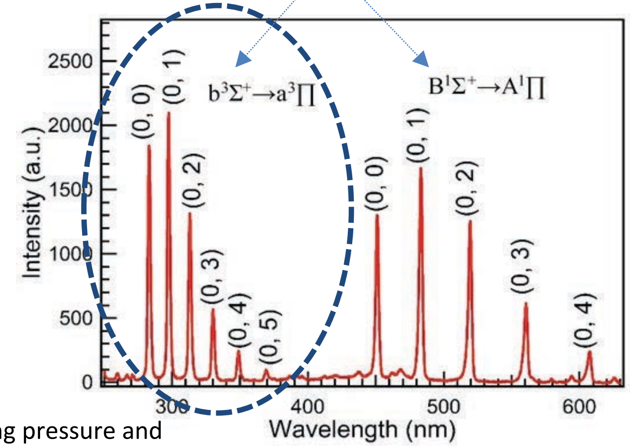
**Ultrashort Pulse: Femtosecond-Duration (1 fs = 10^{-15} s)*

Molecular Species Imaging: Carbon Monoxide (CO) TPLIF

CO TPLIF Excitation/Detection

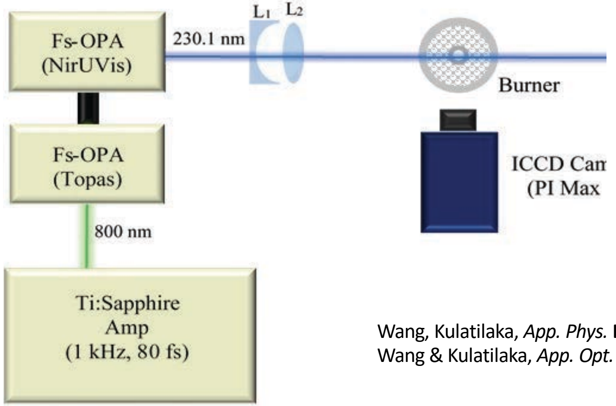


Pure CO in a gas cell at 1 bar



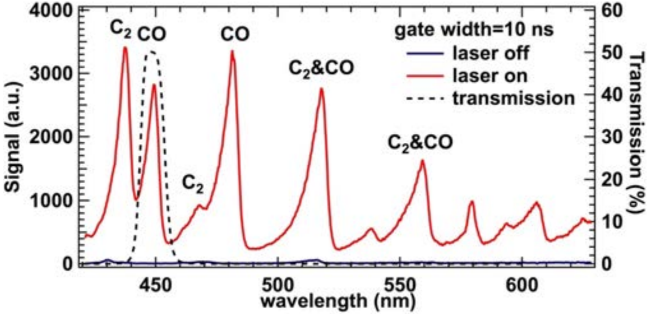
Strong pressure and quenching

CO fs-TPLIF: Experimental



Wang, Kulatilaka, *App. Phys.* **B,124:B** (2018)
Wang & Kulatilaka, *App. Opt.* **58** (2019)

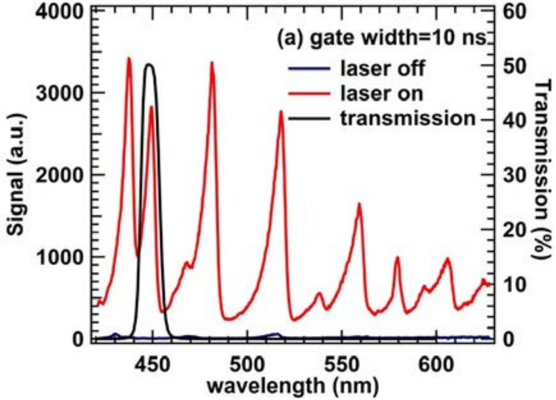
CO TPLIF spectrum in flames: C_2 interreferences



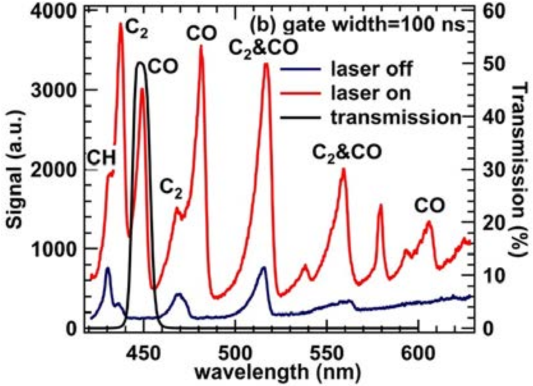
Wang, Jain, Kulatilaka, *Proc. Combust. Inst.*, ***37*** (2019)

C_2 Interferences: Detection Gate Width & Spectral Filtering

ICCD Gate: 10 ns

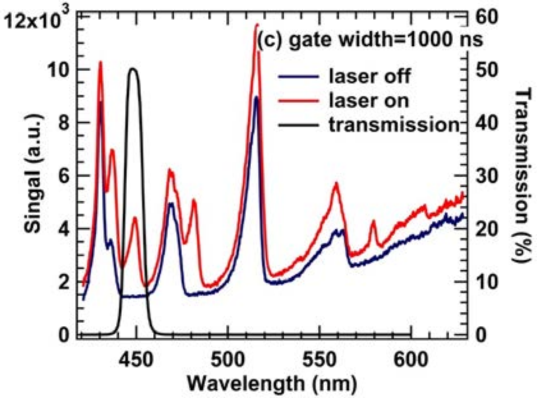


ICCD Gate: 100 ns



Premixed C_2H_4-air flame with equivalence ratio Φ=1.5

ICCD Gate: 1000 ns

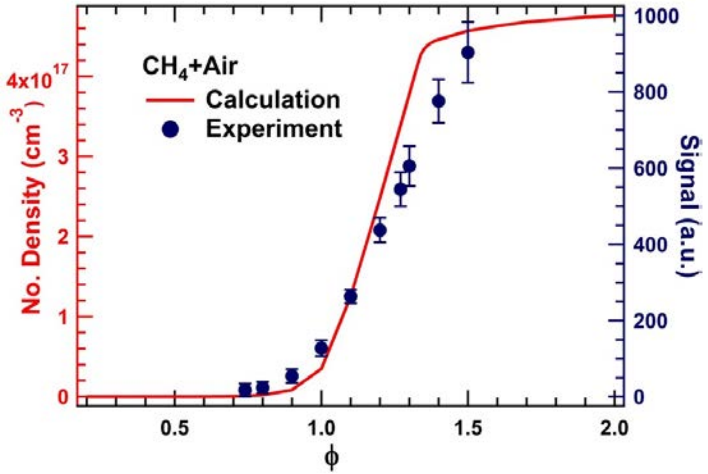


- C_2 cross-talk reduces available CO detection lines
- Narrower detection gate width (2–10 ns) reduces C_2 interference and as well as LIF signal

Wang, Kulatilaka, *App. Phys.* **B,124:B** (2018)

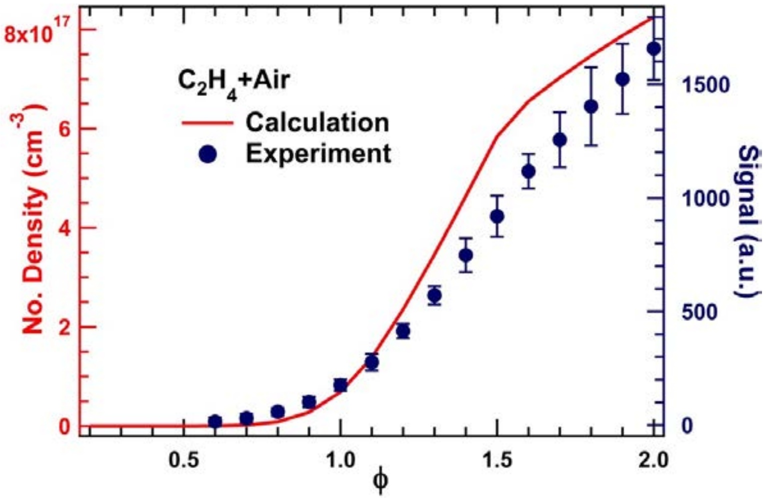
Fs-TPLIF of CO in Methane and Ethylene (Sooting) Flames

Premixed CH_4/Air Flame



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

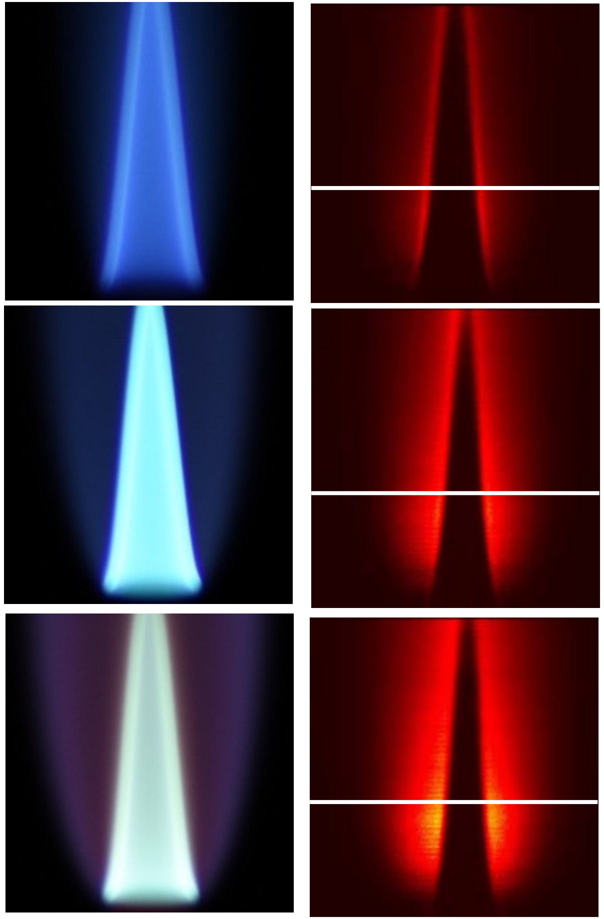
Premixed C_2H_4/Air Flame



Wang, Kulatilaka, *App. Phys.* **B,124:B** (2018)

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

CO Distribution Profiles (shot-averaged)

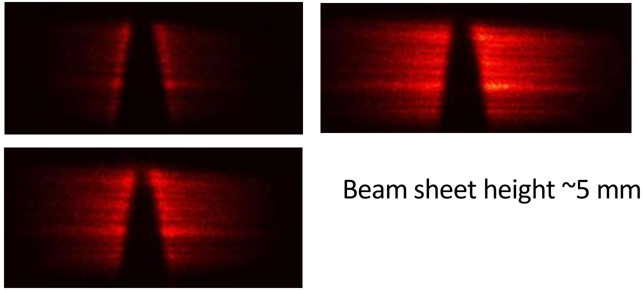


Φ=1.0

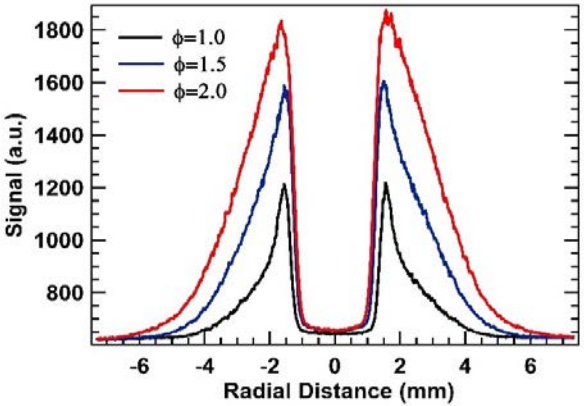
Φ=1.5

Φ=2.0

Single-Shot Images

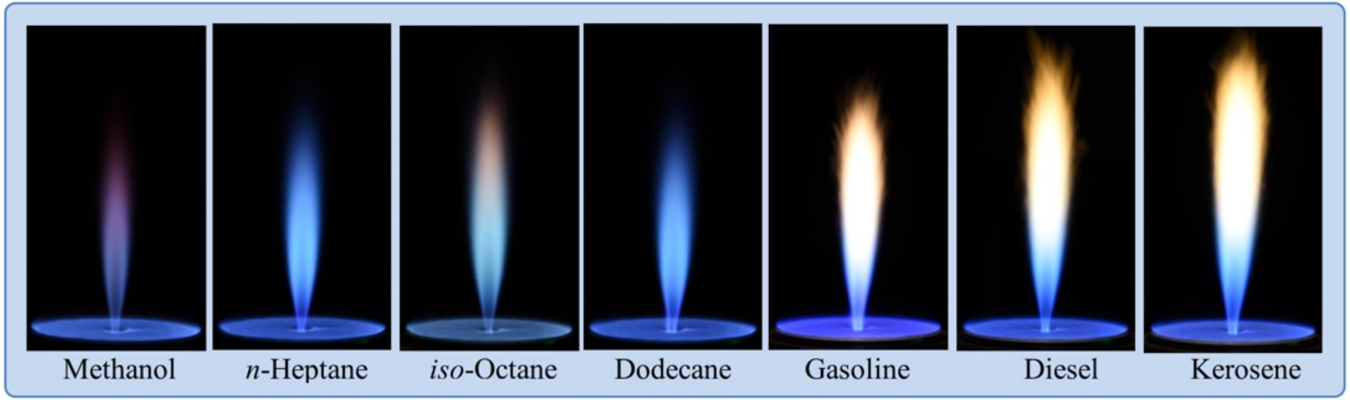


Beam sheet height ~5 mm

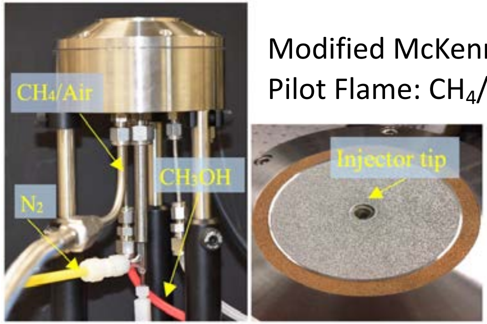


Wang, Kulatilaka, *App. Phys.* **B,124:B** (2018)

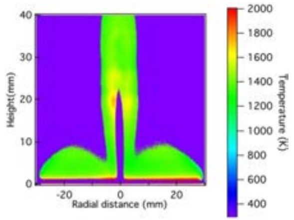
CO Imaging in Piloted Liquid-Fuel Spray Flames



Multi-Fuel Operation

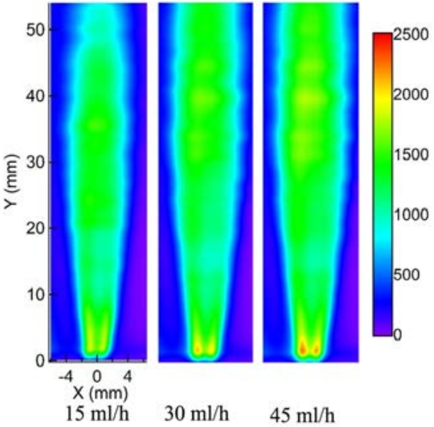


Modified McKenna Burner
Pilot Flame: CH_4/Air



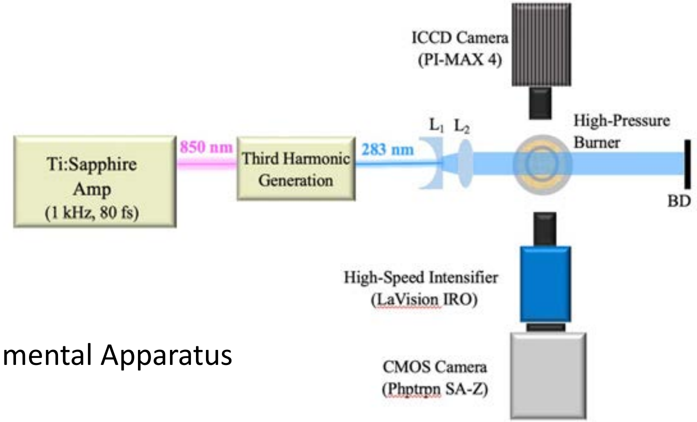
Two-Color OH
PLIF Temp

2D CO Images

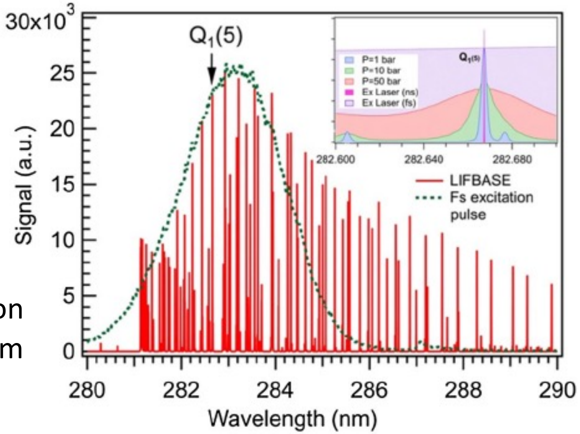


Wang, Kulatilaka, et al., *Combust Sci. Technol.* ***192*** (2019)

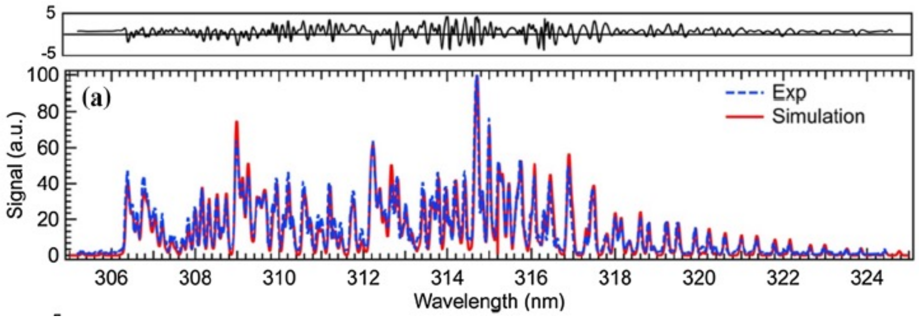
Molecular Species Imaging: Hydroxyl Radical (OH) PLIF *(single-photon excitation)*



Experimental Apparatus

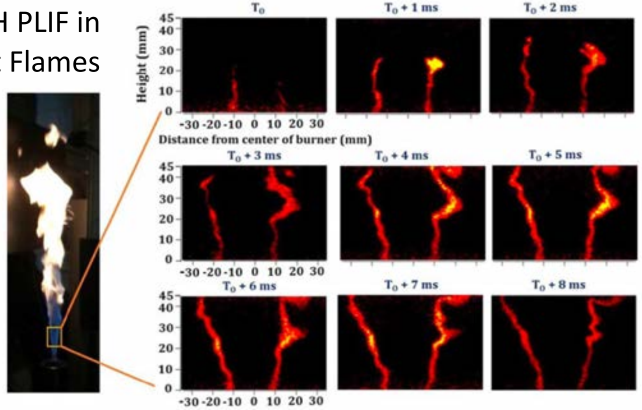


OH Excitation Spectrum



High-Resolution Emission Spectrum

1-kHz OH PLIF in Turbulent Flames



Wang, Kulatilaka et. al., *App. Phys. B,* **125:90** (2019
Wang, Kulatilaka et. al., *Comb. Flame,* **214** (2020)

Webinar Outline

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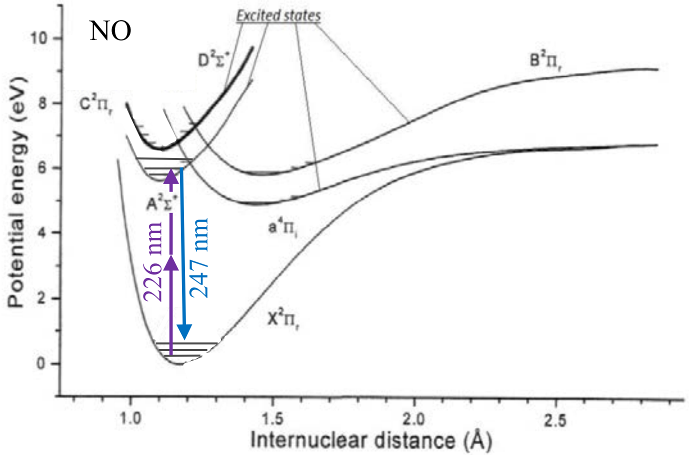
III. Molecular Species Imaging: *CO & OH*

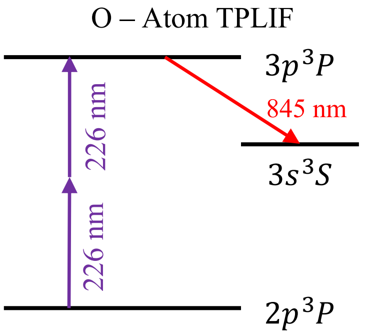
IV. Simultaneous Multi-Species Imaging Using a Single Femtosecond Laser *(H/OH, NO/O/O_2)*

**Ultrashort Pulse: Femtosecond-Duration (1 fs = 10^{-15} s)*

Excitation and Detection Scheme for NO, O, & O_2

- NO excitation/detection
  - NO A-X (0,0) excited at 226 nm. Reduce O_2 interferences at this wavelength
  - Strong emission at 236- and 247-nm bands
- O excitation
  - $3p^3P \leftarrow\leftarrow 2p^3P$ two-photon transition at 226 nm
  - 845-nm emission from $3p^3P \rightarrow 3s^3S$ decay
- O_2 excitation
  - $B^3\text{–}X^3\Sigma_g^-$ Schumann-Runge system
  - Highly temperature dependent
  - Excited by laser radiation from 175–535 nm
  - Various emission lines from 200–400 nm

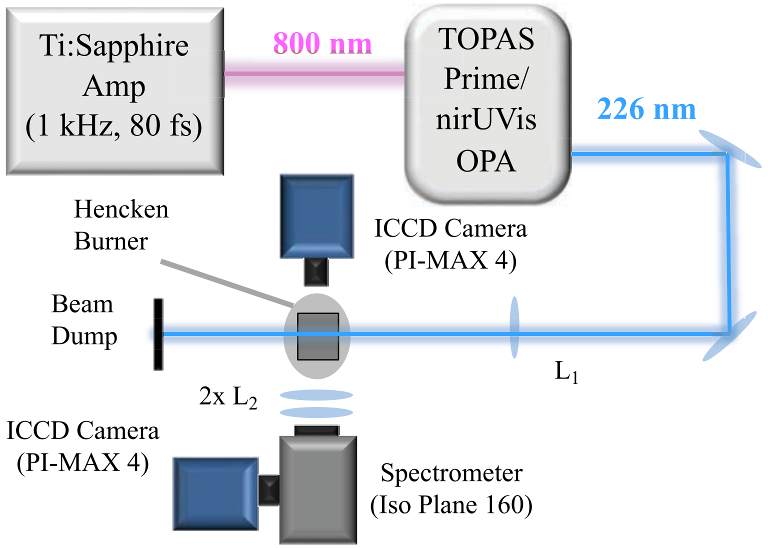




W.G. Bessler, C. Schulz, T. Lee, J.B. Jefferies, R.K. Hanson,, Appl. Opt. 42 (2003) 4922-4936.

Experimental Apparatus for Simultaneous NO, O, & O_2 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 O_2 line images
  - Direct Imaging with visible ICCD Camera for single-shot O- atom detection

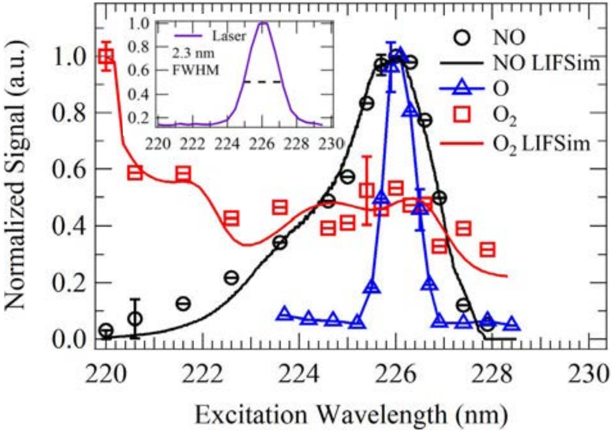


Hay, Kulatilaka, et al., *Proc. Combust. Inst.* In Press. (2023)

NO, O, & O_2 Excitation and Detection Scans

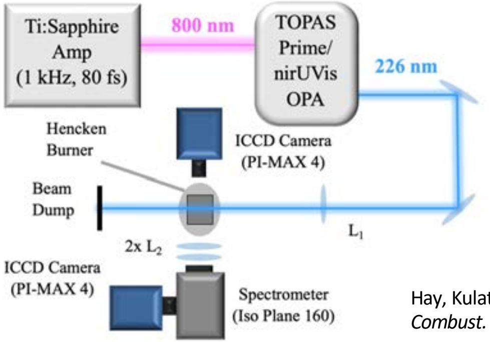
- Laser excitation wavelength scan:
  - OPA output wavelength scanned from 220–228.5 nm
  - 226.1 nm radiation maximizes NO and O-atom signal. Still produces some O_2 excitation
  - NO and O_2 experimental data compared to convolutions of LIFSIM simulations

- Laser energy scan:
  - Excitation wavelength fixed at 226.1 nm
  - Linear energy dependence on laser energy for NO and O_2
  - Quadratic dependence on laser energy for O-atom



CH_4 – Air Bunsen Burner

H_2 – Air Hencken Burner



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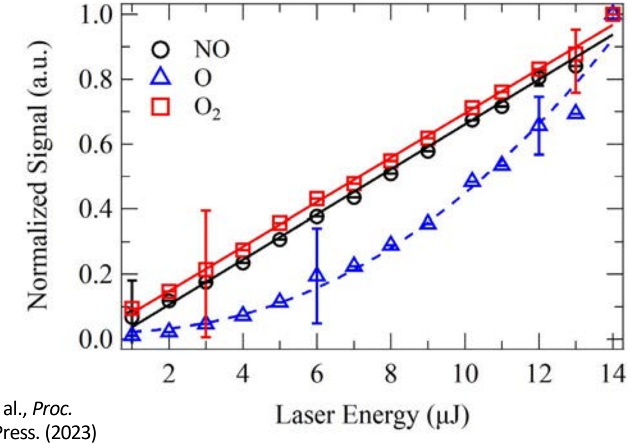


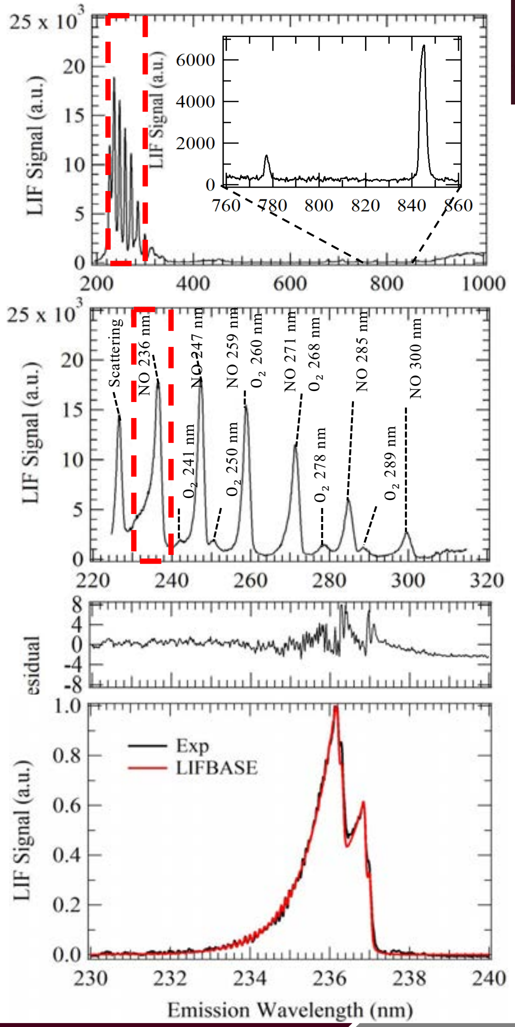
NO, O, & O_2 Detection Spectral Characterization

- NO, O, and O_2 LIF signals collected through an imaging spectrometer (300 lines/ mm and 1200 lines/mm gratings)

- Interference-free emission lines for NO and O_2:
  - NO: 236, 247, and 285 nm
  - O_2: 241, 250, 278, and 289 nm

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

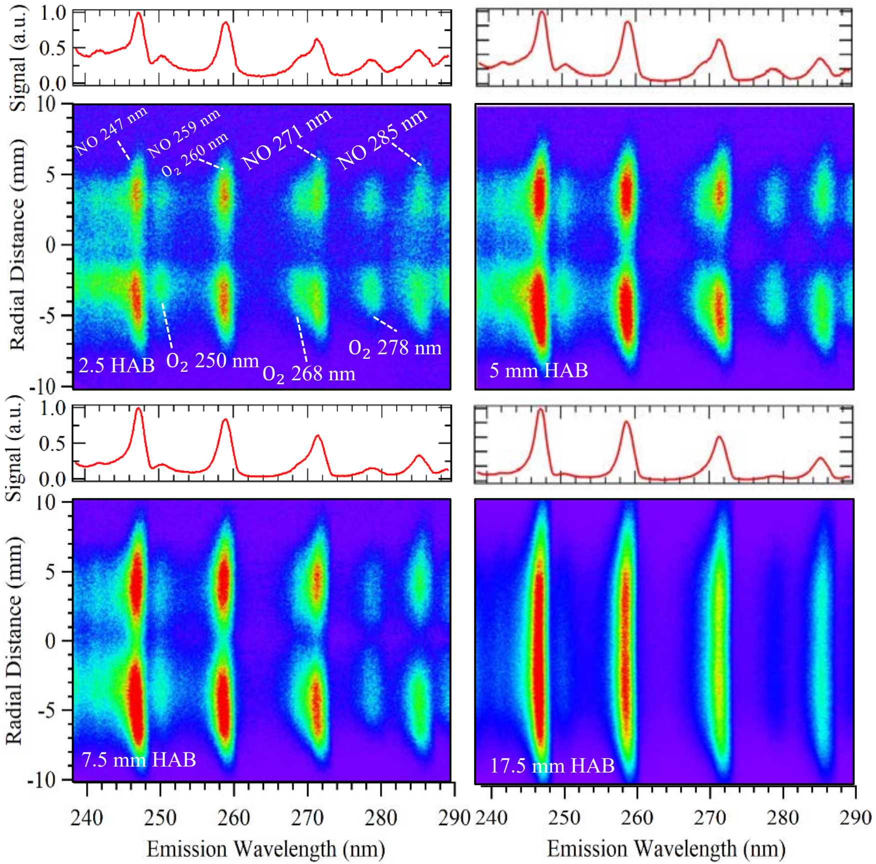
- Estimated NO detection limit between 10~40 ppm in flames

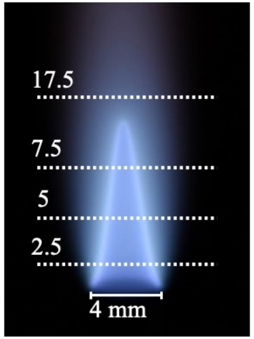


Hay, Kulatilaka, et al., *Proc. Combust. Inst.* In Press. (2023)

NO, O, & O_2 Detection Spectral Characterization – 1D Imaging

- NO and O_2 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). O_2 decreases as it is consumed by the flame

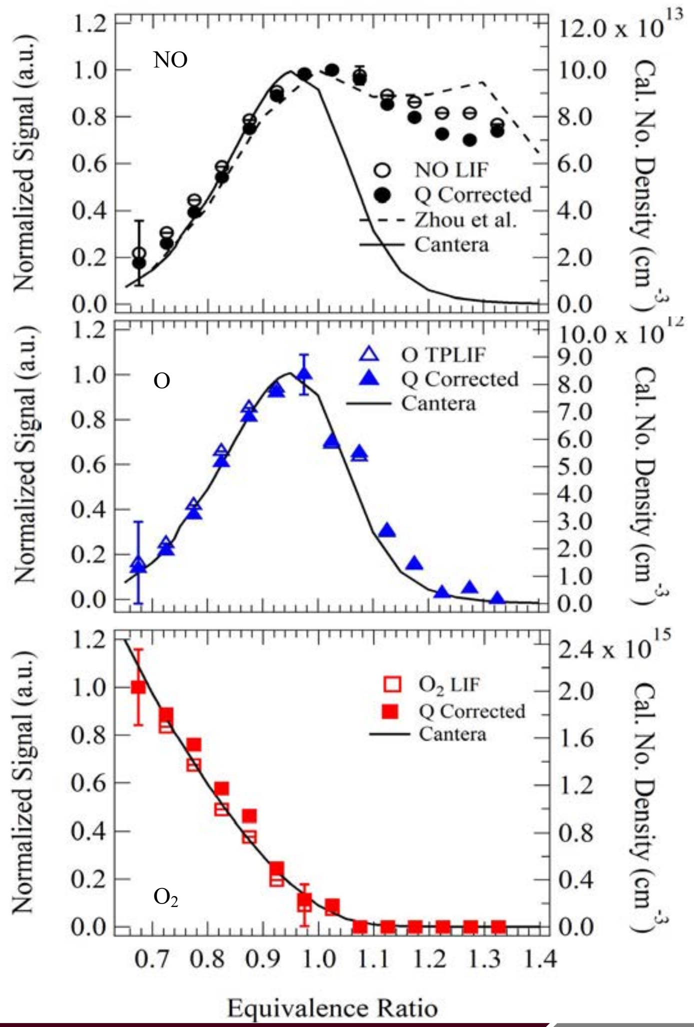




Hay, Kulatilaka, et al., *Proc. Combust. Inst.* In Press. (2023)

NO, O, & O_2 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, N_2, H_2, CH_4, H_2O, CO_2, and O_2 as colliders
- Departure in rich NO measurements is likely due to prompt NO formation



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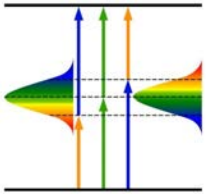
In Summary,

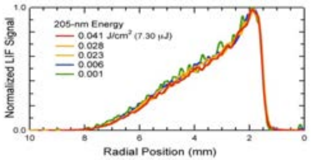
Ultrashort Pulse Laser Diagnostics for Chemically Reacting Flows

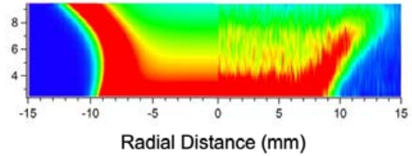
I. Some Basics of Femtosecond Pulses



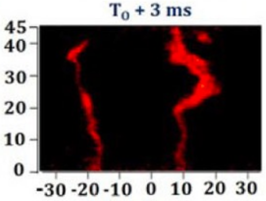
II. Fs-TPLIF of Atomic Species: *Basics & Some Applications*

fs excitation (~100 fs)

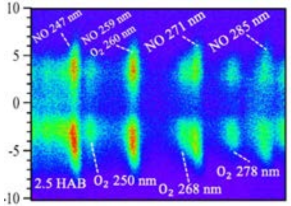




III. Molecular Species Imaging: *CO & OH*



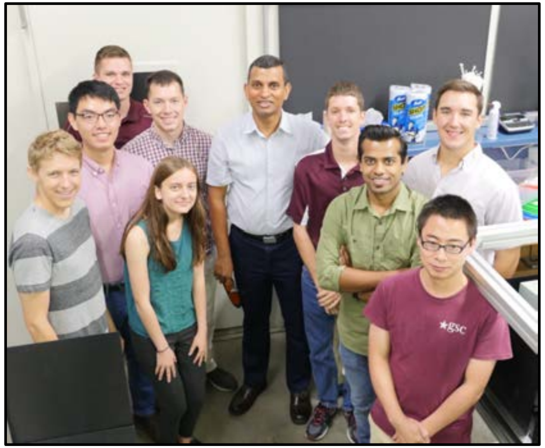
IV. Simultaneous Multi-Species: *NO/O/O$_2$*



Acknowledgements

Current & Former Members:

- Morgan O’Neil
- Yejun Wang
- Ayobami Shoyinka
- Tyler Pascal
- Nicholas Niemiec
- Ayush Jain
- Will Swain
- Pradeep Parajuli
- Christian Schweizer
- Gavin Lukasik
- Matt Hay
- Kristi Naude
- Andrew Gorman
- Matthew Intardonato
- Manuel Suarez



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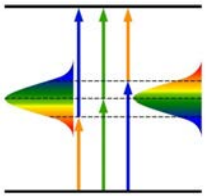

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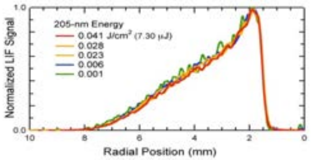

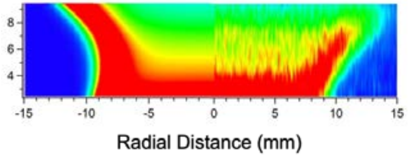

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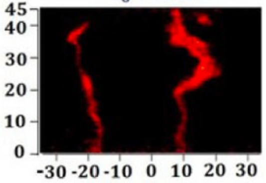

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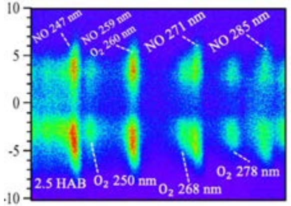

Q & A