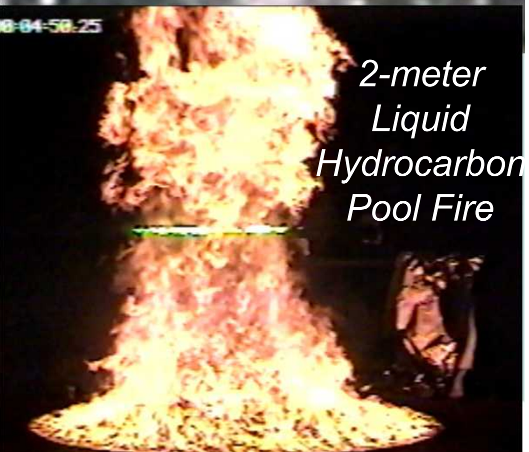

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Exceptional service in the national interest

SAND2015-9944PE

CARS Diagnostics in Hostile Combustion Environments

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U.S. DEPARTMENT OF ENERGY

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Preview

- Background
  - Sandia (NM) applications in “hostile” environments
  - CARS fundamentals (nanosecond)
- 2-meter pool fire study with nanosecond laser pulses
- Ultrafast (fs/ps) CARS development
  - Why ultrafast?
  - “Hybrid” rotational CARS
  - Canonical flame measurements
- Applications of ultrafast CARS: sooting and particle-laden flames
  - Sooting turbulent jet flame
  - Solid carbon-epoxy composite fuel
  - Aluminized propellant
- Pressure measurements (bonus material)

Contributions from .....

- Chris Kliewer and Alexis Bohlin
  - 1-D Line CARS measurements at Sandia/CA
  - Etalon design and selection for fs/ps CARS

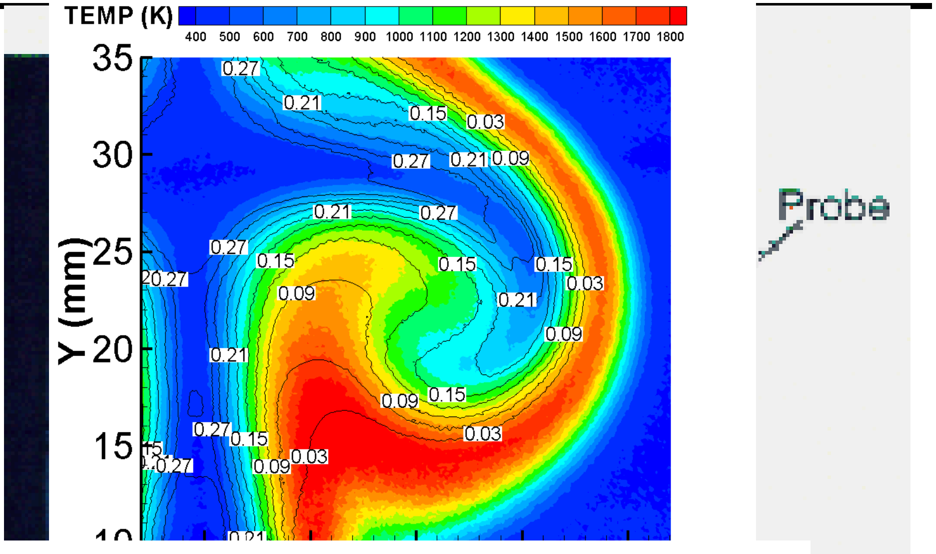
- John Hewson and David Lignell (BYU)
  - One dimensional turbulence modeling of sooting jet flame

- Dan Guildenbecher
  - Digital holography/propellants

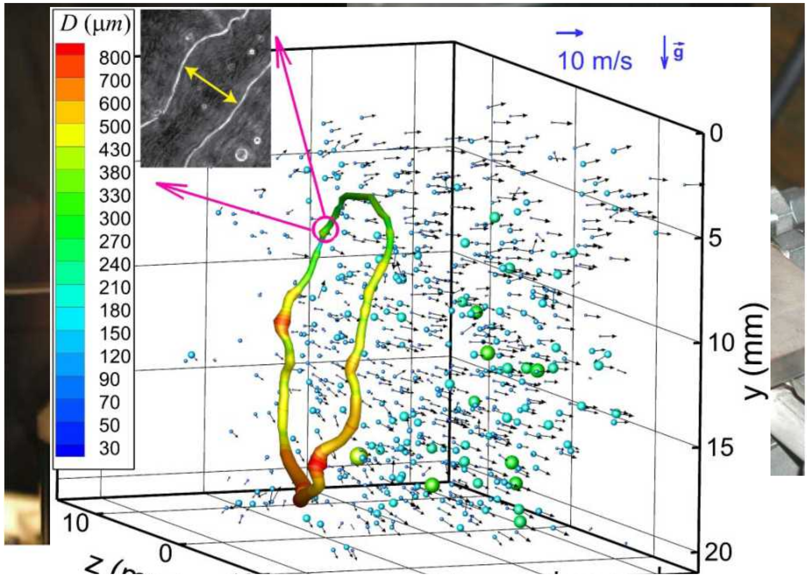
Laser-based diagnostics empower combustion research

- Non-perturbing
- Free of radiation and insertion errors
- 2-D or even 3-D ***quantitative*** imaging
- Multiple parameters (T/species/soot/velocity…)

- High temporal resolution – 10 ns or better
- High spatial resolution – 10^{-4} – 10^{-5} cm^3
- Most effective in clean, laboratory flames



Kearney et al., *Applied Optics* **44** (2005)

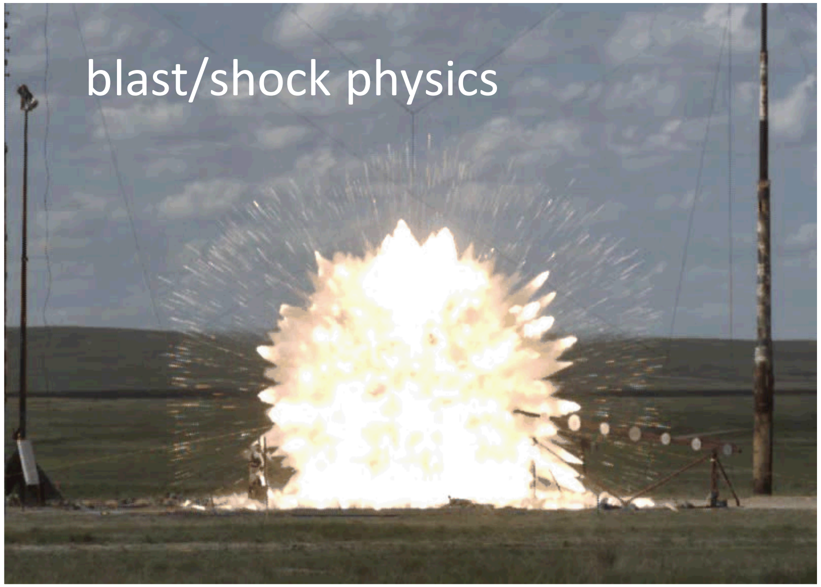


Gao et al., *Optics Letters* **38** (2013)

Sandia/NM National Security Mission Space

- ***DOE strategic systems safety***
- ***DoD/WFO applications***
- ***Challenging environments***
- ***Large-scale***
- ***Heat, blast, particulate***

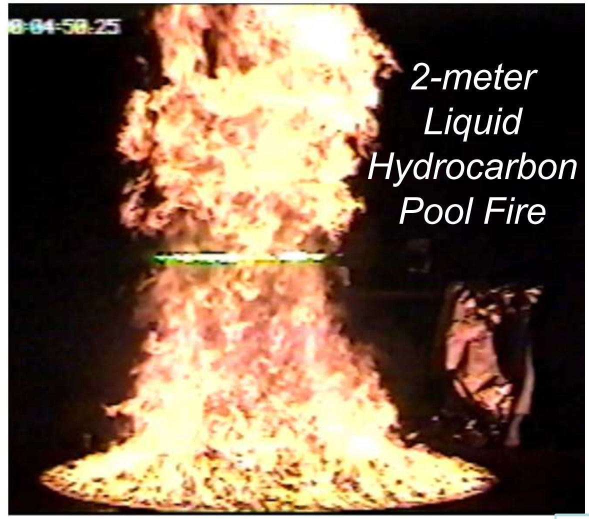
http://www.cbsnews.com/news/rocket-crash-no-immediate-threat-to-station-but-cause-is-unknown/

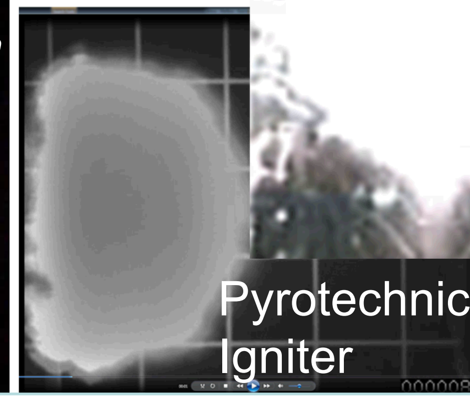


http://abcnews.go.com/Technology/Travel/wireStory?id=3529012

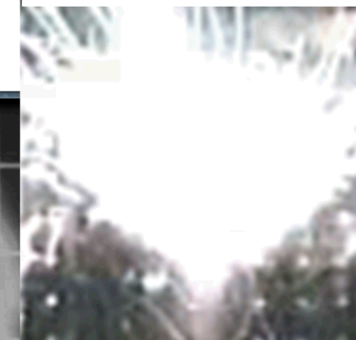
Sandia's application space presents significant challenges

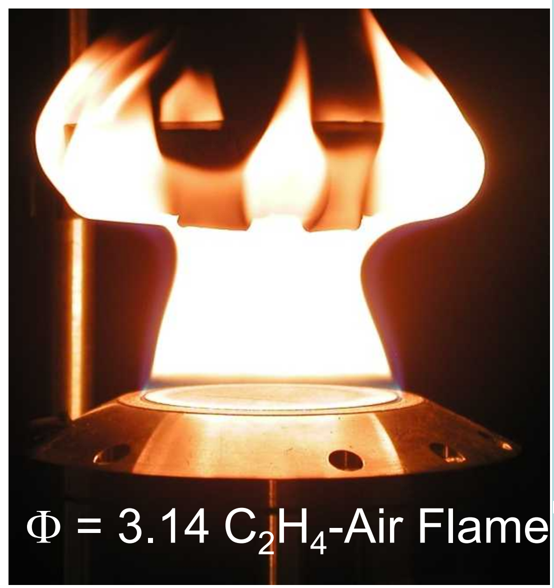
- "Dirty" environments
  - Fire research
  - Energetic materials
- Soot, aluminum particulate
- Luminosity
- Scattering
- Absorption/optical thickness
- ***Large-scale*** of combustion systems

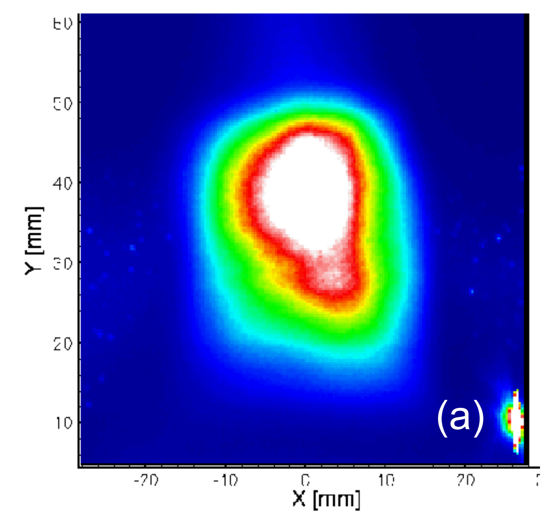




Propellant Burn

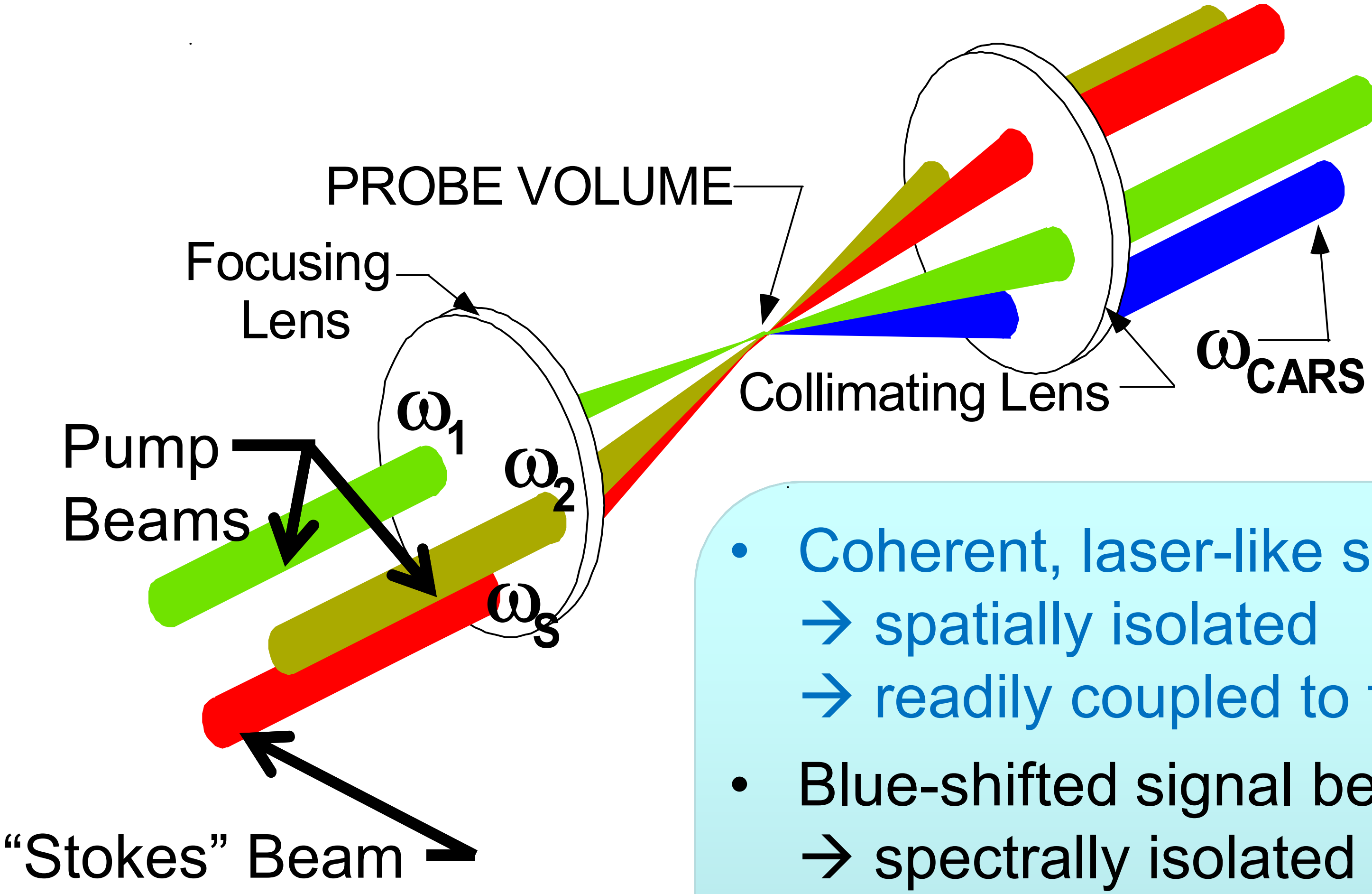






Sooting

Coherent anti-Stokes Raman Scattering (CARS)



- Coherent, laser-like signal beam
  → spatially isolated
  → readily coupled to fibers
- Blue-shifted signal beam
  → spectrally isolated
- Orders of magnitude stronger than incoherent scattering

CARS Physical Processes: Light/Matter Interaction

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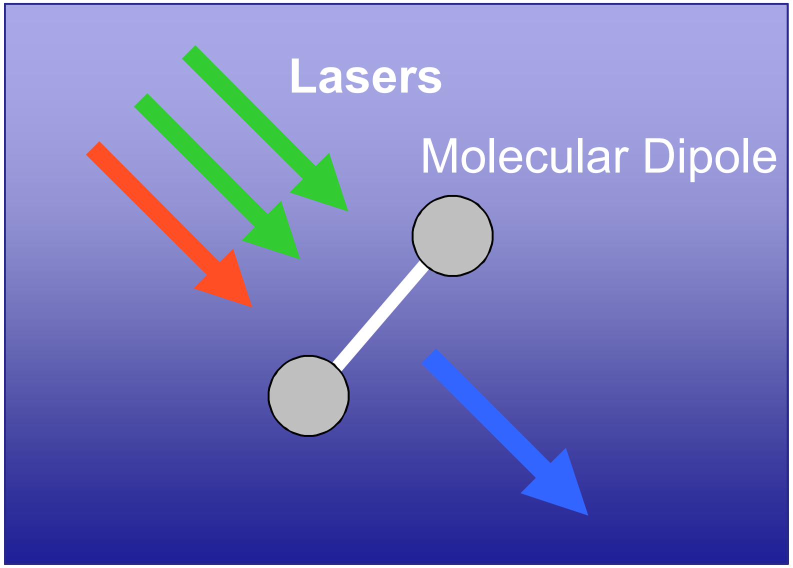
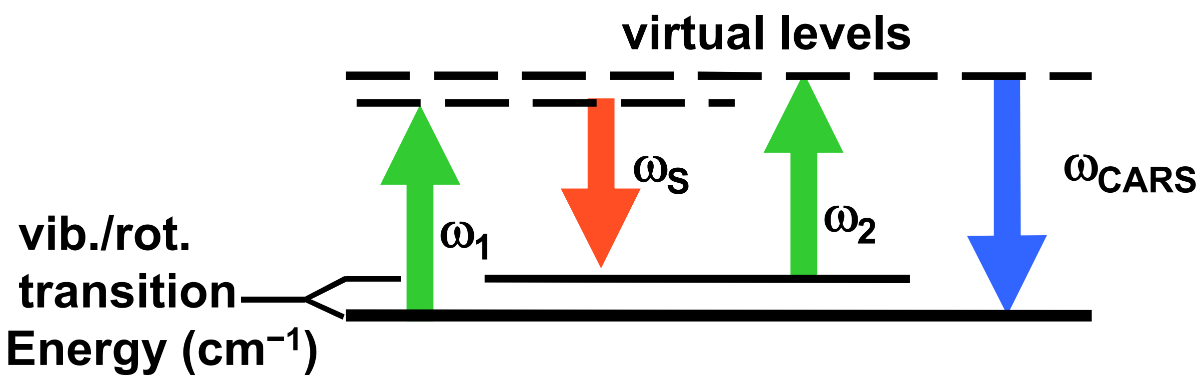
- A 'polarization' or induced dipole is prepared by pump and Stokes beams
- This polarization scatters the second pump wave
- Constructive interference in one *phase-matched* direction only

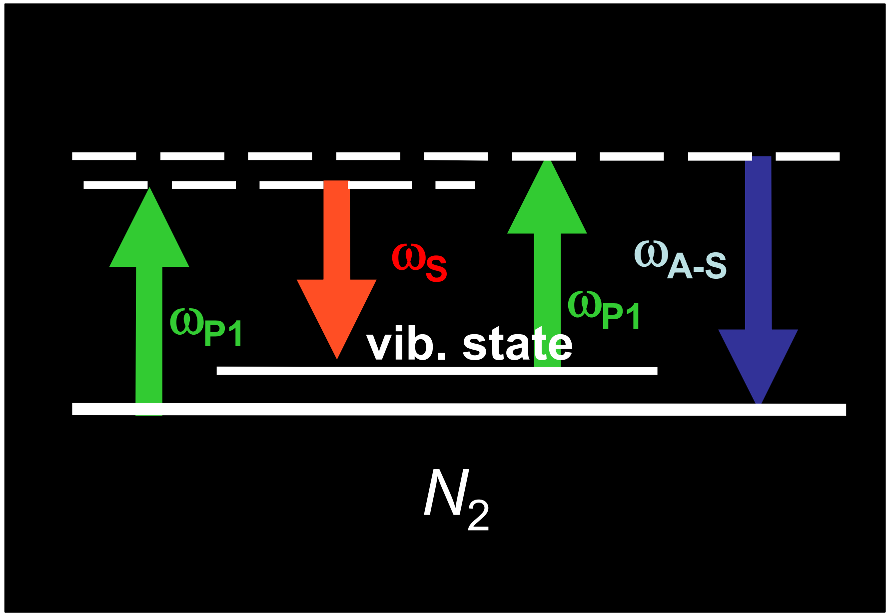


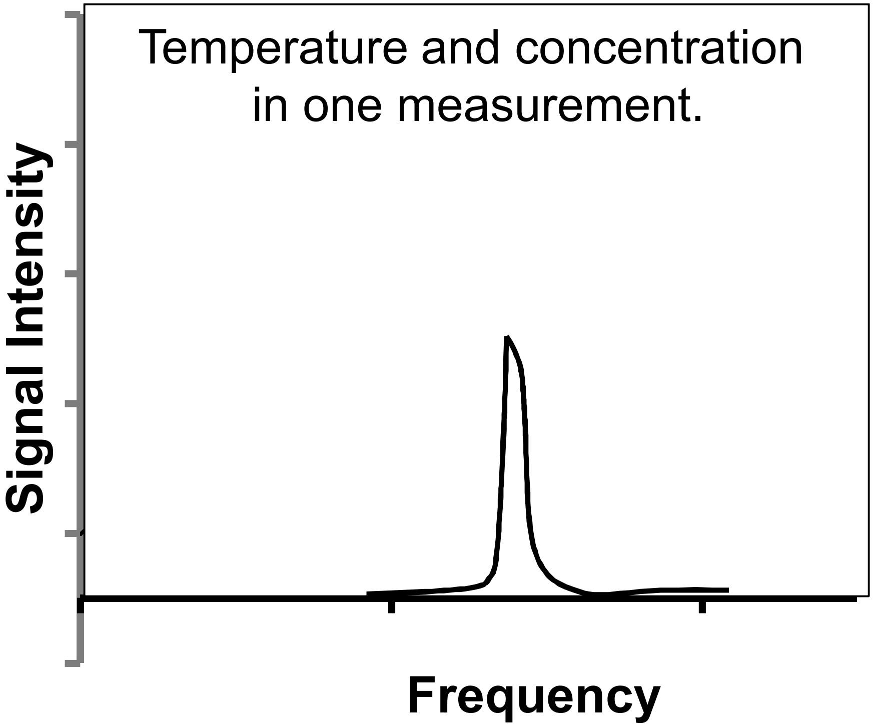
Coherent Anti-Stokes Raman



A broadband source permits single-shot detection

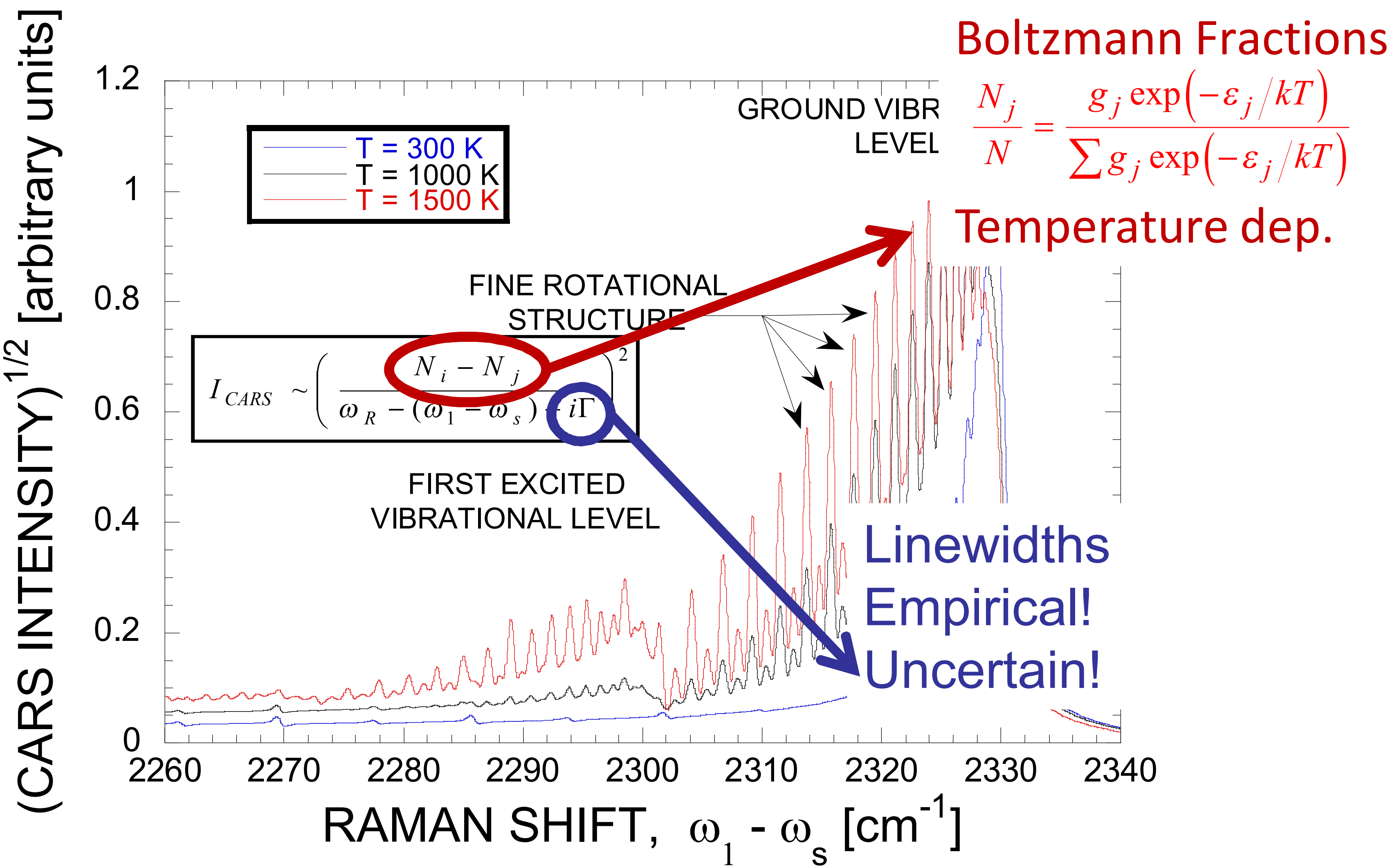
- If all lasers are narrowband one energy level is probed





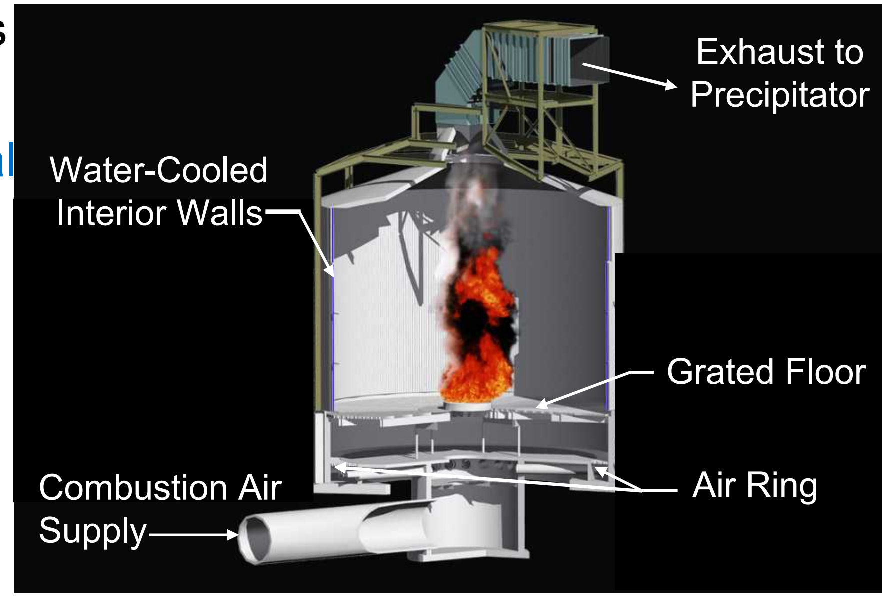
- If one (or more) laser is broadband then a range of energy levels differences are probed

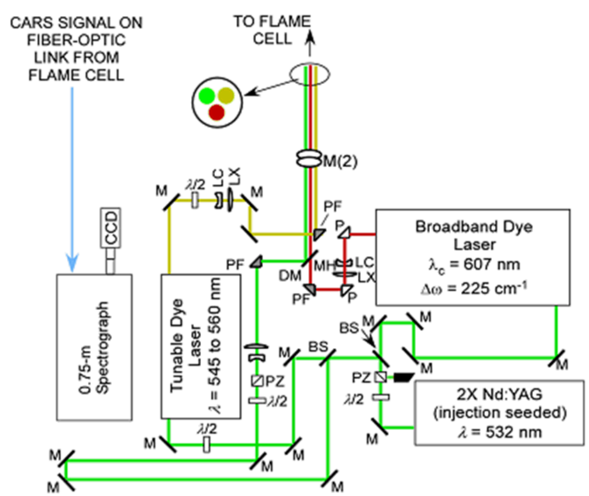
Temperature sensitivity comes from the spectral shape

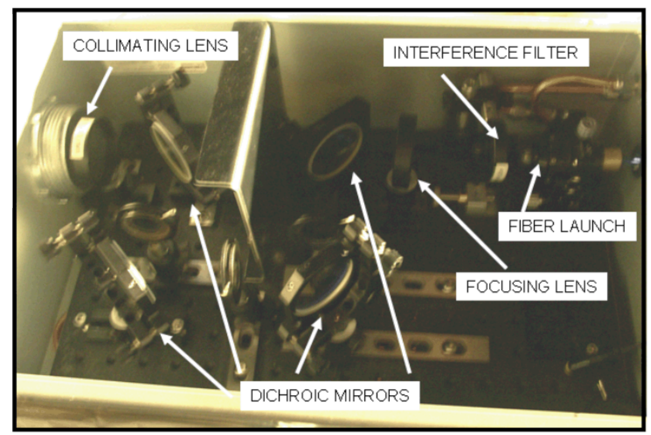


CARS Probing of 2-m Diameter Liquid Pool Fires

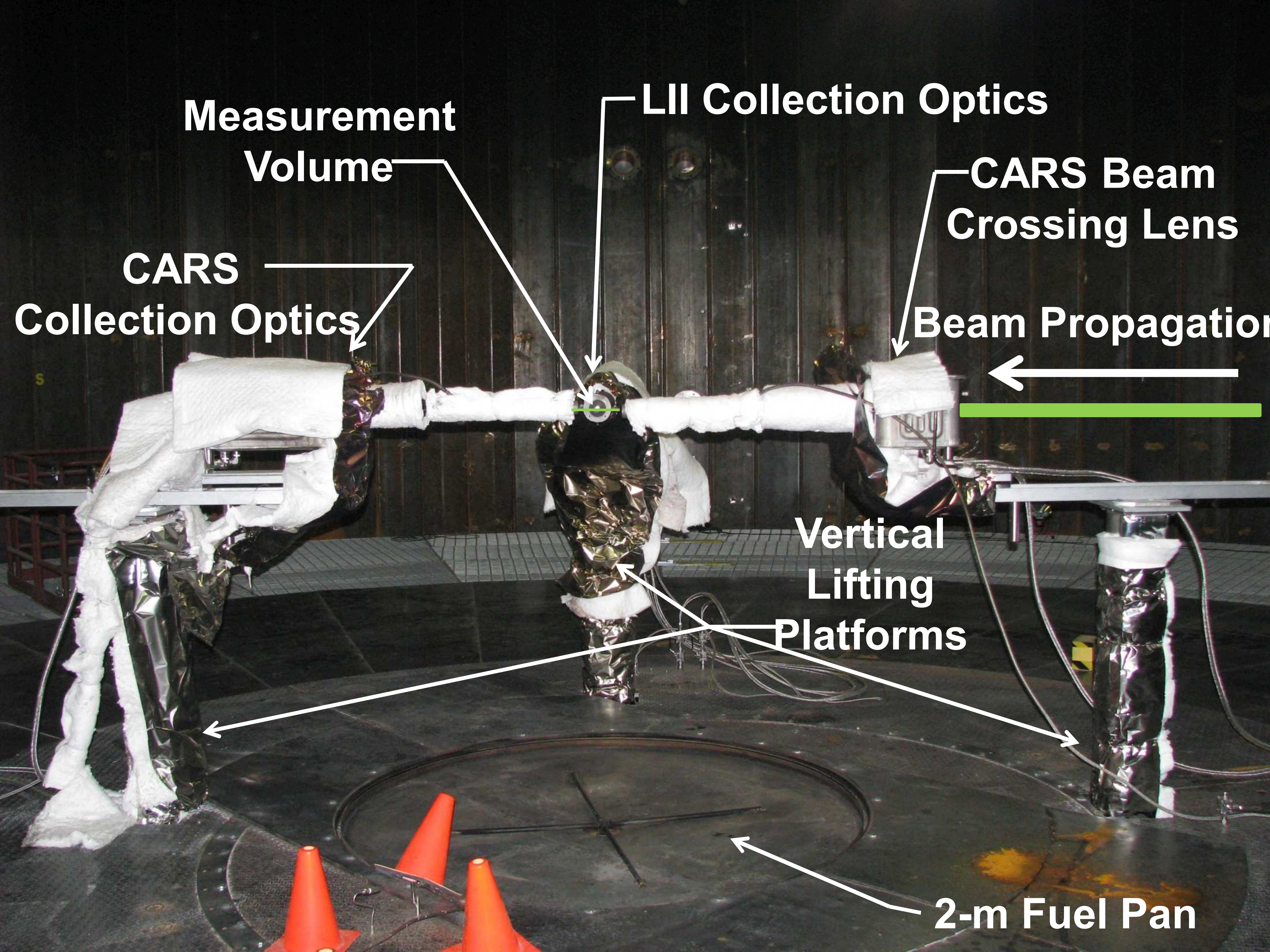
- Fire Laboratory for Accreditation of Models and Experiments (FLAME) Facility
- Designed to facilitate deployment of optical diagnostics for full meter-scale fire testing
- Laser laboratories with optical access on three sides
- Large scale positioning system to move optical and pool fire
- Fiber-coupled CARS (and soot LII) diagnostics fielded





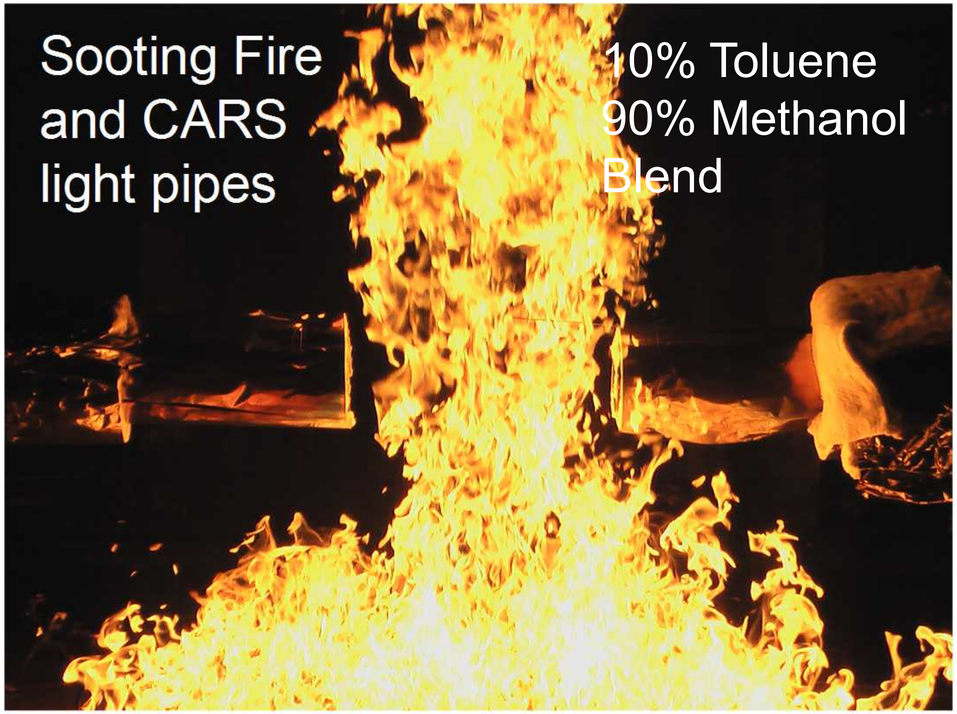


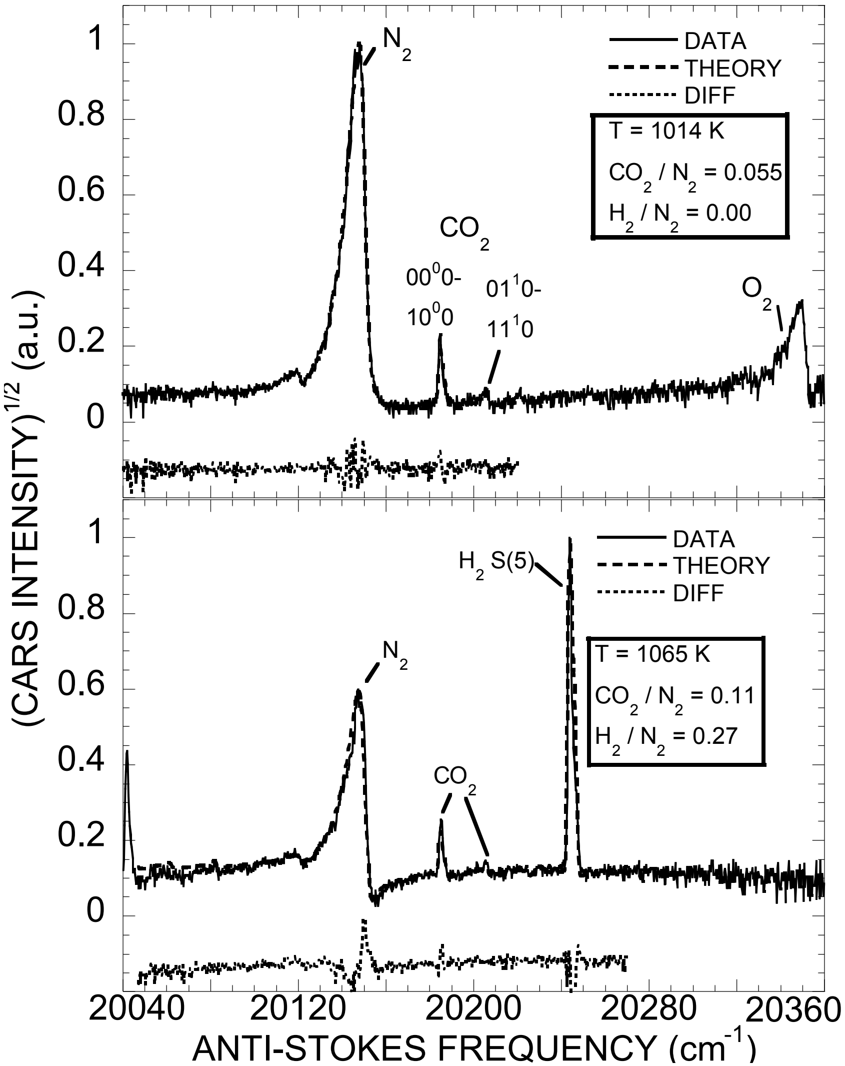
Water-cooled enclosure
Flame engulfed optics!


Measurement
Volume
LII Collection Optics
CARS Beam
Crossing Lens
CARS
Collection Optics
Beam Propagation
Vertical
Lifting
Platforms
2-m Fuel Pan

Single-Shot Spectra Provide Simultaneous Temperature/Species Information in Sooting Fire

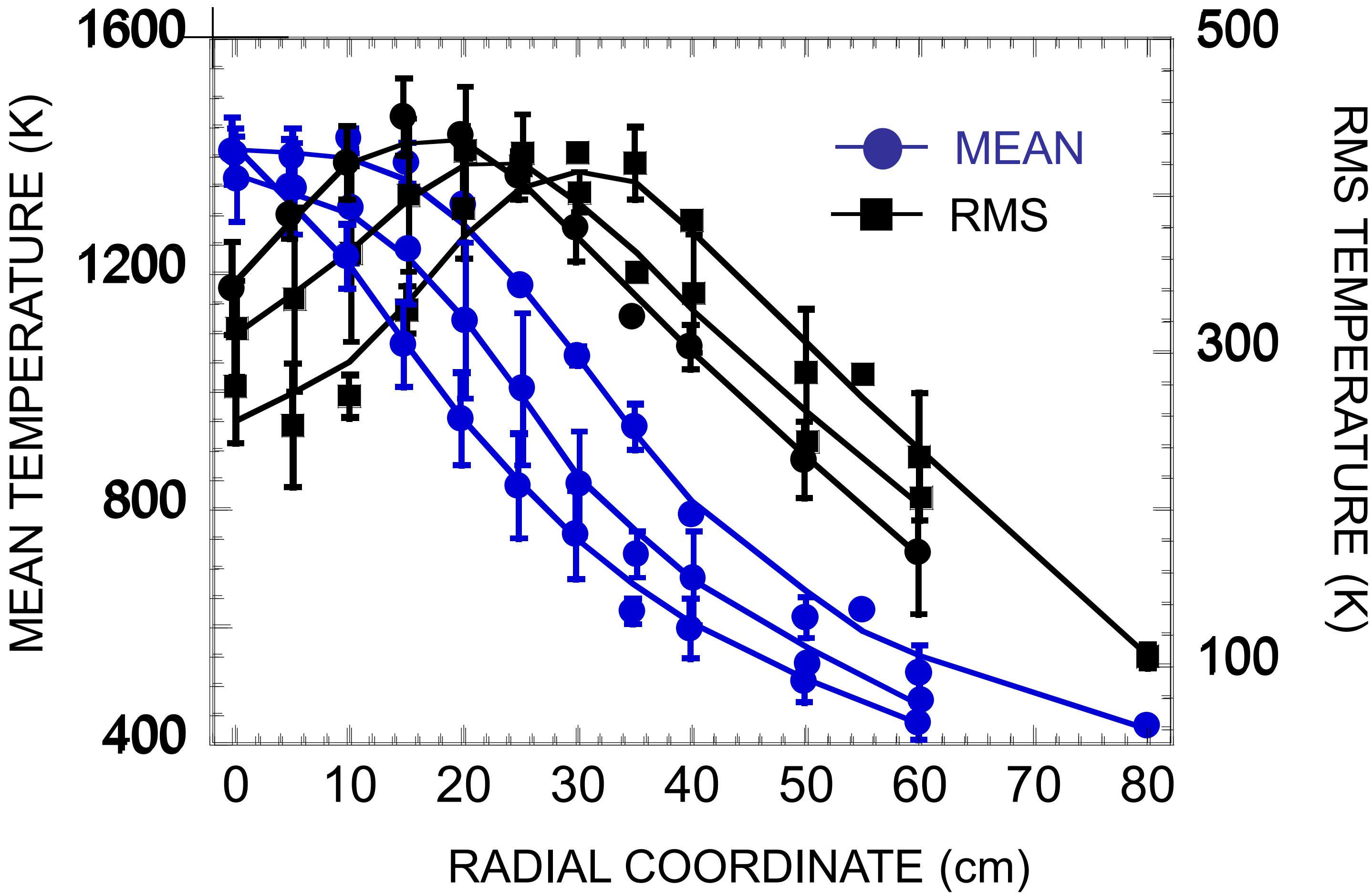
- CARS spectra from sooting fire show N_2, CO_2, H_2, and O_2
- Full ensemble of species data not available as of yet
- Two representative spectra with theoretical fits (Sandia CARSFT code) shown here



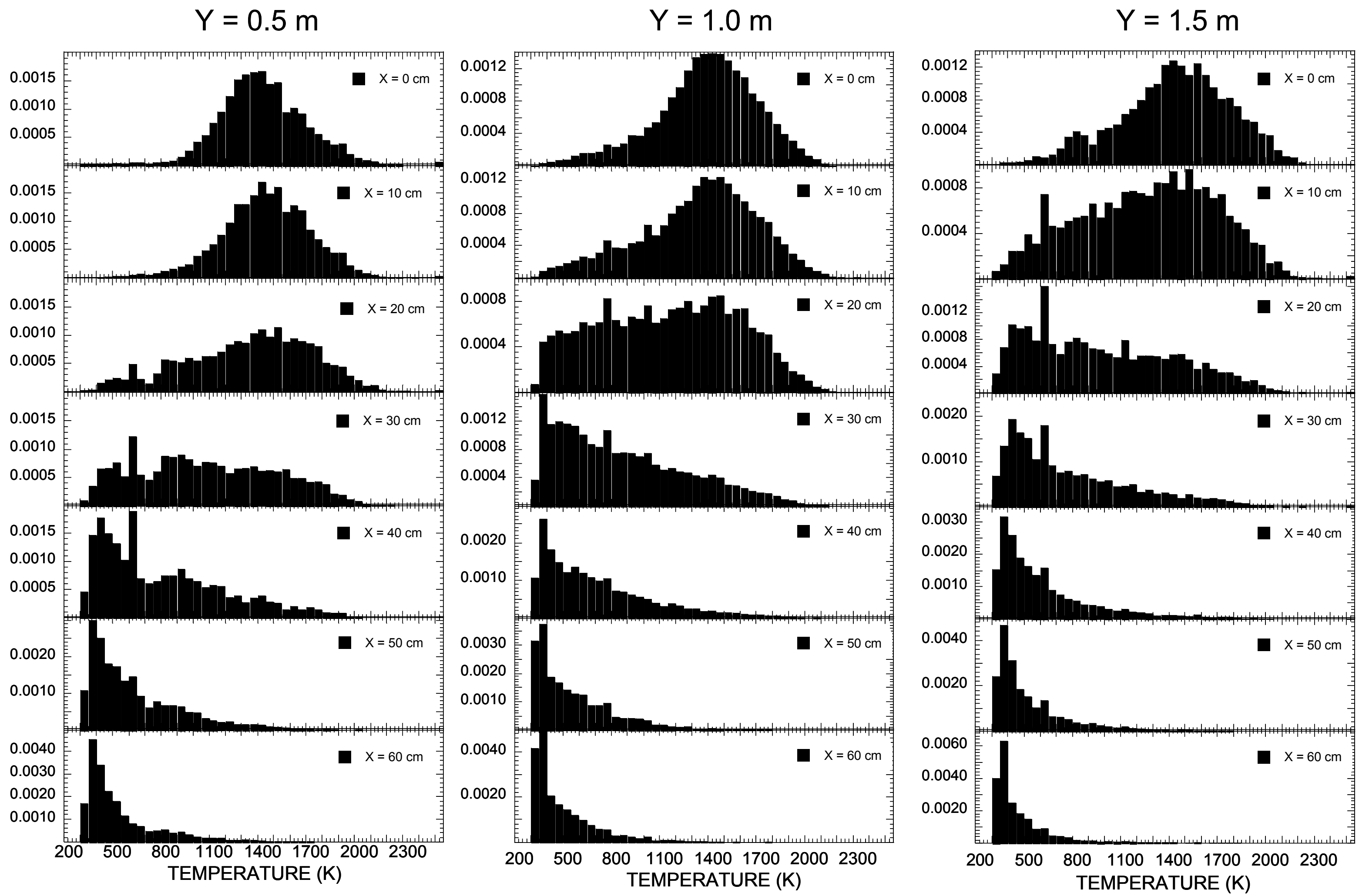


Kearney *et al.*, Proc. Combust. Inst. **32**, 871-878, 2009.

Radial Temperature Profiles – Sooting Pool Fire



Temperature PDF – Sooting Pool Fire



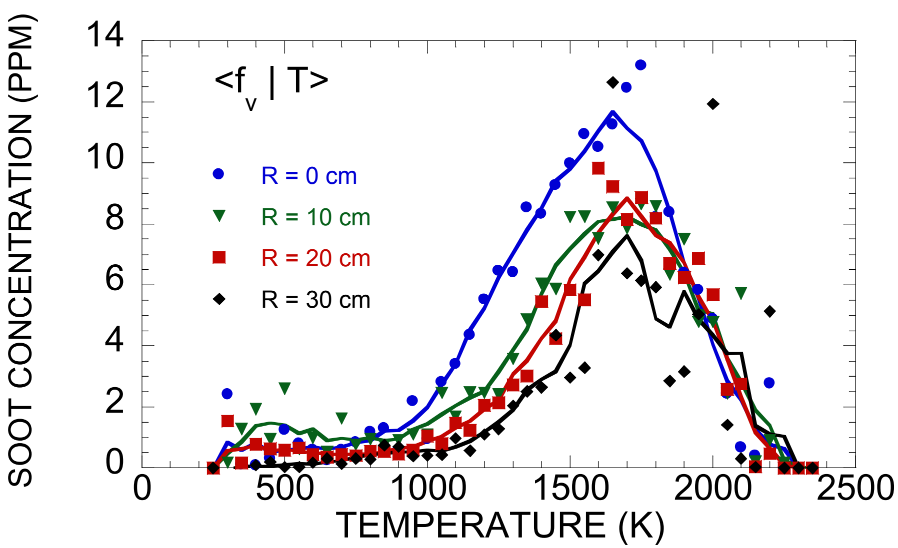
Joint/Temperature Soot Statistics for Emission

Radiative Transfer Equation

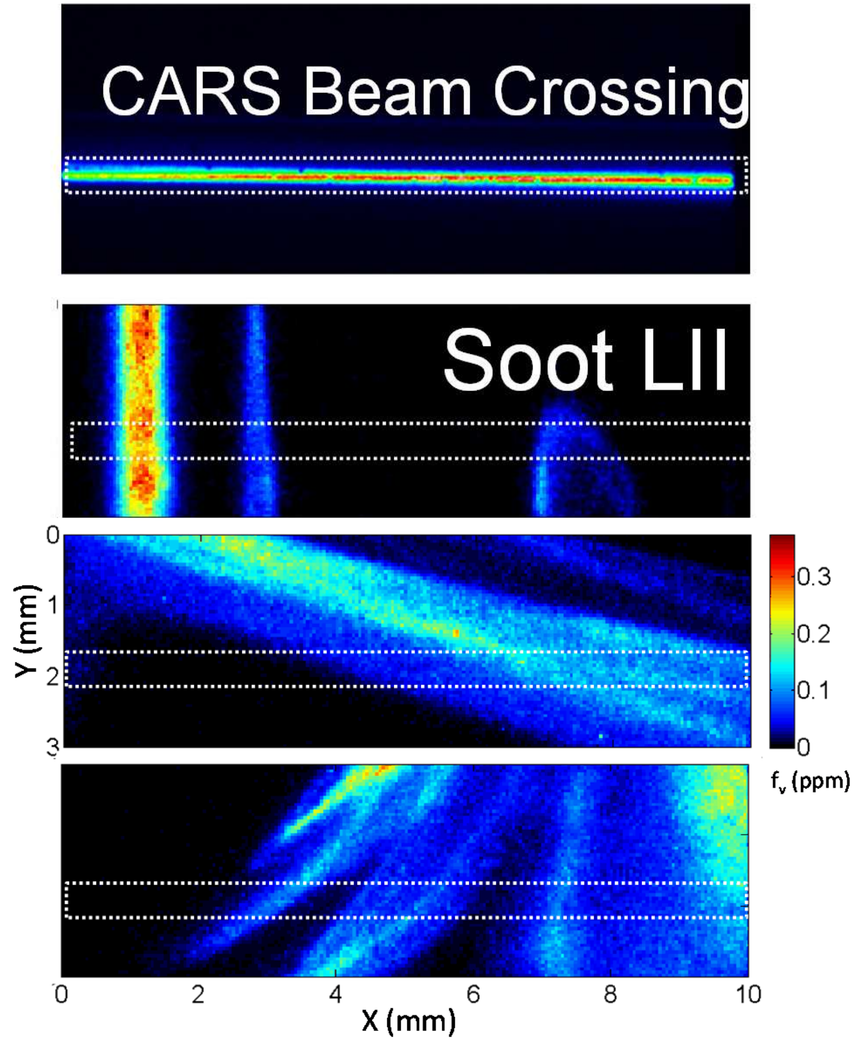
$$\frac{d\overline{I_\lambda}}{ds} = \overline{\mu_\lambda I_{\lambda,b}(T)} - \overline{\mu_\lambda}\,\overline{I_\lambda}$$

Joint Temp. Soot Statistics Desired

- CARS system combined with LII soot detection
- Average soot in 10^{-5} cc CARS volume correlated with enthalpy-pooled temperature



Mean Soot Conditioned on Temperature



Frederickson and Kearney, Appl. Opt. **50** (2011)
Kearney and Pierce, Combust. Flame **159** (2012)

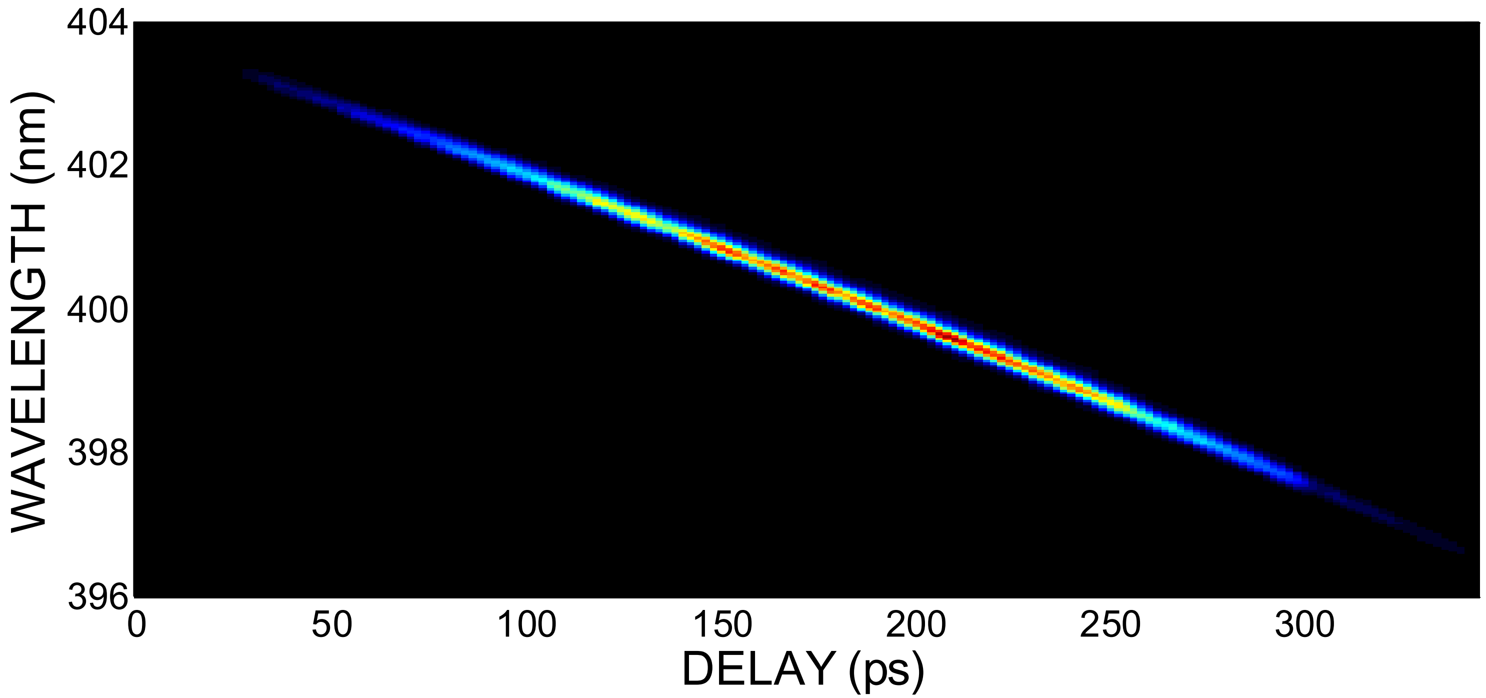
Femtosecond CARS

Why Ultrafast?

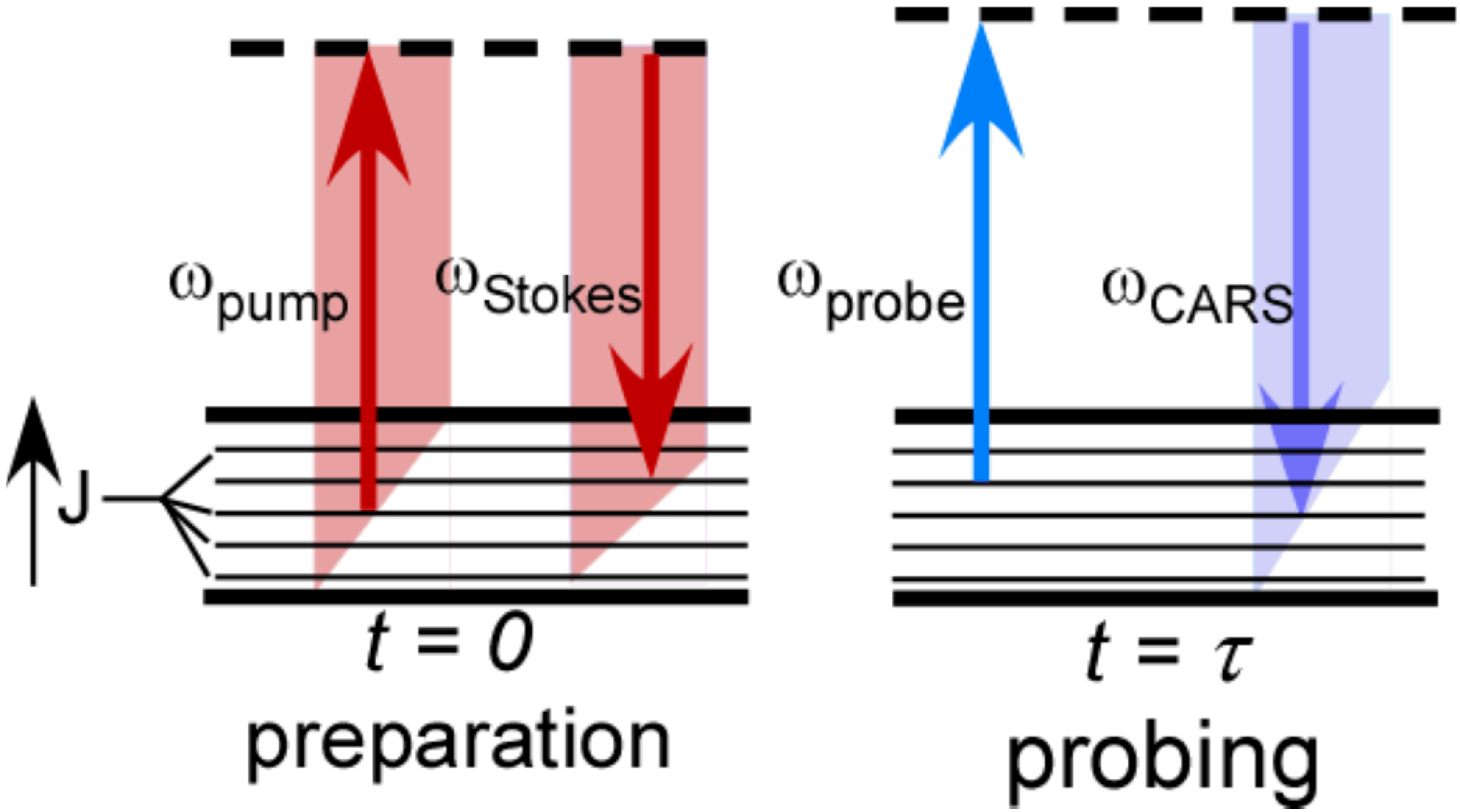
- High-quality (transform-limited) ***broadband*** sources

$$\Delta\tau \, \Delta\nu \geq const.$$

- High repetition rates (kHz vs 10 Hz)
- Transient vs. steady state measurements → no linewidths!
- Bandwidth manipulation



Time-Domain Rotational Raman: $\tau_{laser} << \tau_{molecule}$



"*The story here is really in the time domain*"

Previous Ultrafast CARS Development

- Purdue faculty (Lucht, Meyer) were among the first to apply ultrafast lasers to gas-phase combustion

APPLIED PHYSICS LETTERS **89**, 251112 (2006)

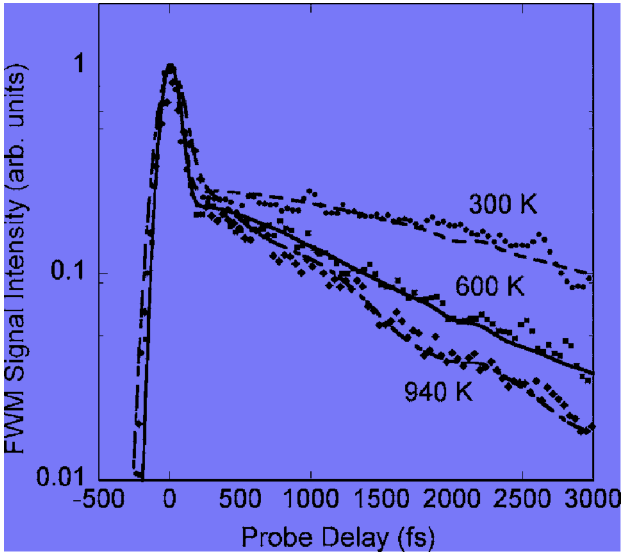
Femtosecond coherent anti-Stokes Raman scattering measurement of gas temperatures from frequency-spread dephasing of the Raman coherence

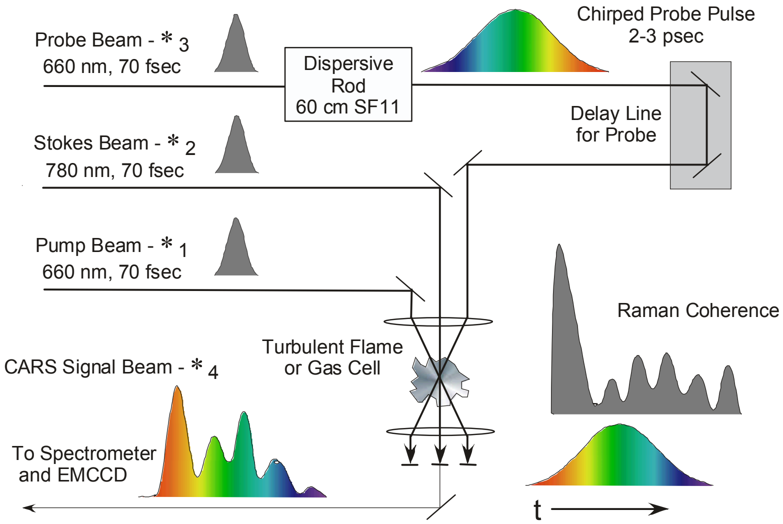
Robert P. Lucht[a)]
School of Mechanical Engineering, Purdue University, West Lafayette, Indiana 47907

Sukesh Roy
Innovative Scientific Solutions, Inc., 2766 Indian Ripple Road, Dayton, Oklahoma 45440

Terrence R. Meyer
Department of Mechanical Engineering, Iowa State University, Ames, Iowa 50011

James R. Gord
Air Force Research Laboratory, Propulsion Directorate, Wright-Patterson AFB, Oklahoma 45433





“Hybrid” Rotational CARS Approach

Single-shot gas-phase thermometry using pure-rotational hybrid femtosecond/picosecond coherent anti-Stokes Raman scattering

Joseph D. Miller,[1] Sukesh Roy,[2] Mikhail N. Slipchenko,[3] James R. Gord,[4] and Terrence R. Meyer,[1,5,*]

[1]Department of Mechanical Engineering, Iowa State University, Ames, Iowa 50011, USA
[2]Spectral Energies, LLC, Dayton, Ohio 45431, USA
[3]Weldon School of Biomedical Engineering, Purdue University, West Lafayette, Indiana 47907, USA
[4]Propulsion Directorate, Air Force Research Laboratory, WPAFB, Ohio 45433, USA
[5]Erlangen Graduate School in Advanced Optical Technologies (SAOT), Friedrich-Alexander University Erlangen-Nürnberg, Germany
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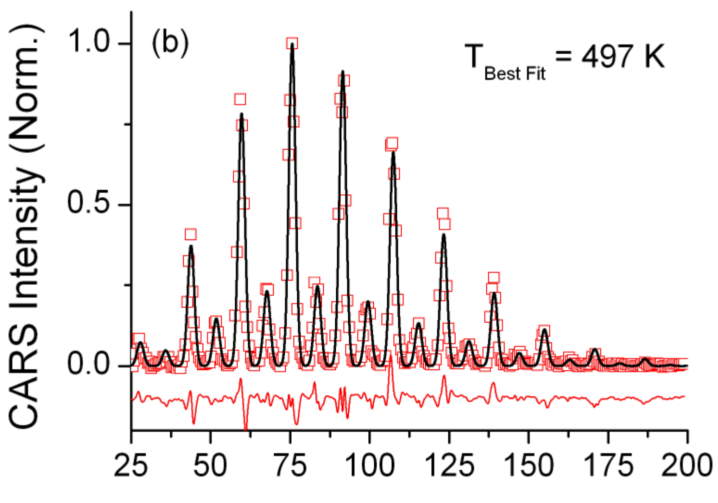
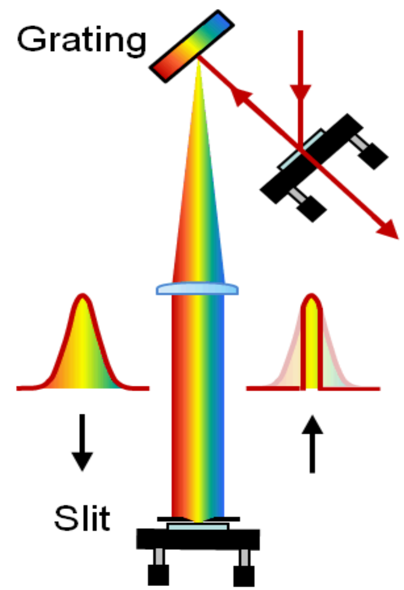
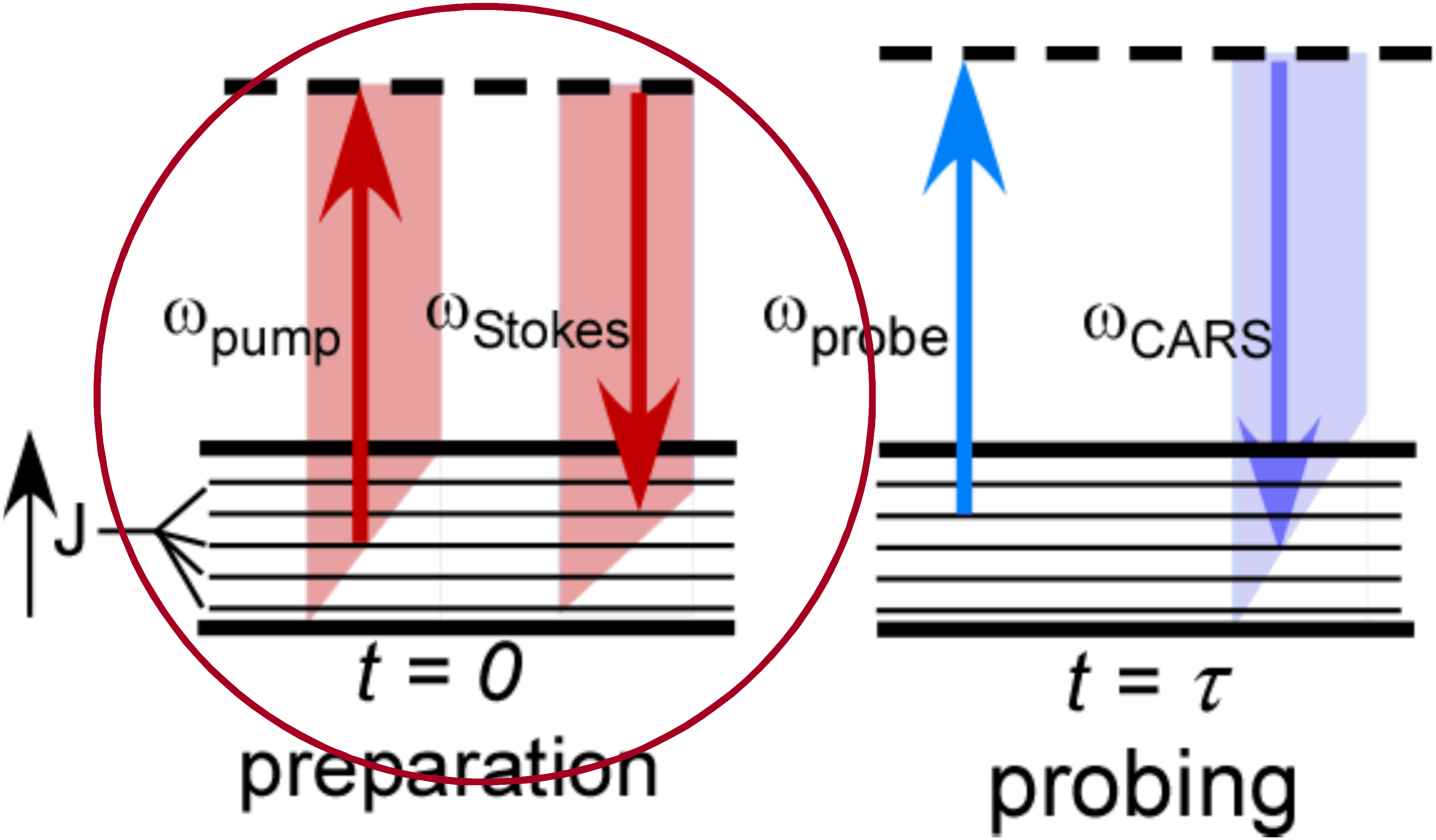
1 August 2011 / Vol. 19, No. 16 / OPTICS EXPRESS 15627

- ***Rotational Raman***
- ***“Hybrid” fs pump/ps probe***
- ***Bandwidth “carving” using a grating-based pulse shaper***
- ***Raman lines are spectrally resolved***



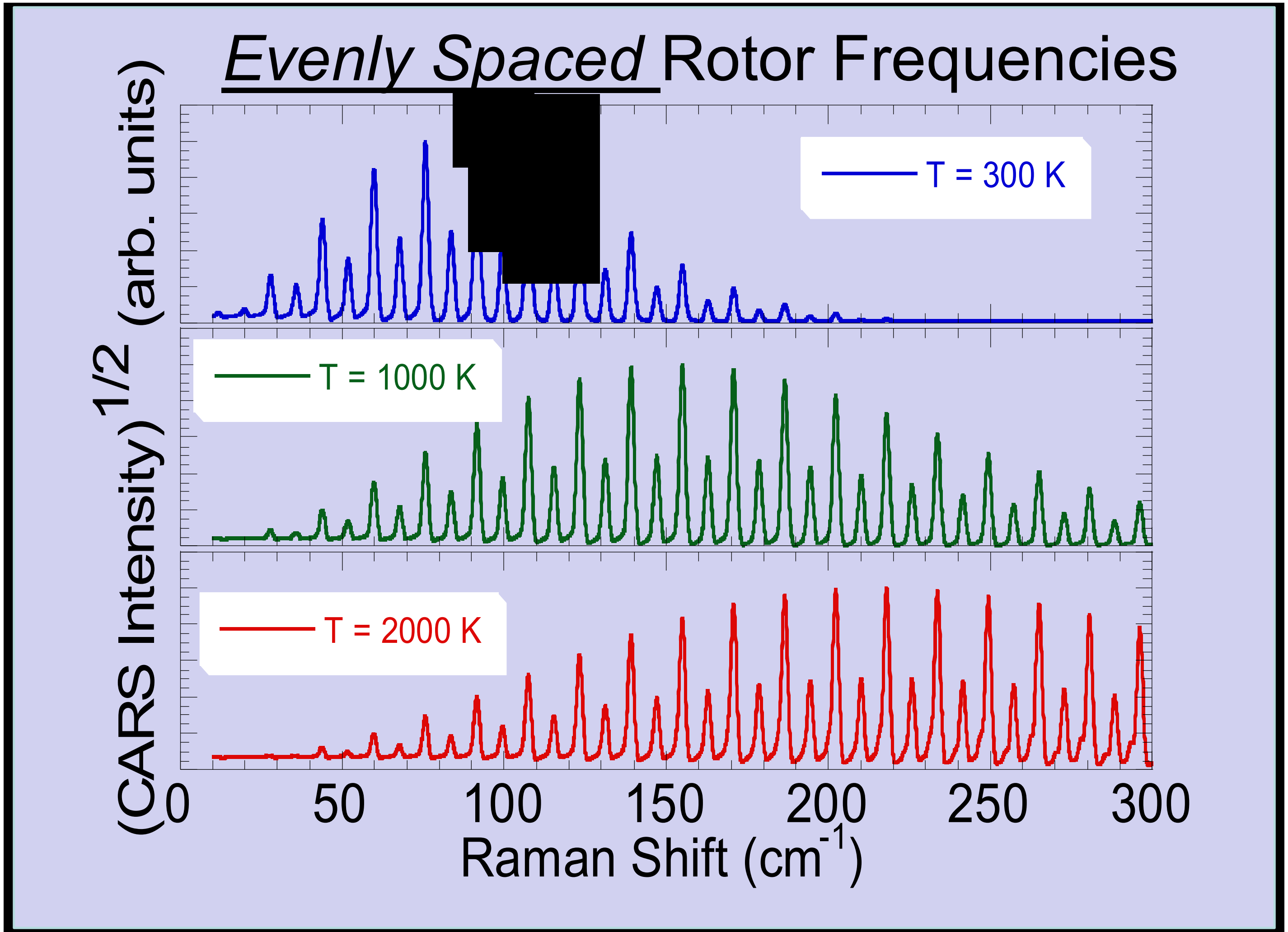


Time-Domain Rotational Raman: $\tau_{laser} << \tau_{molecule}$

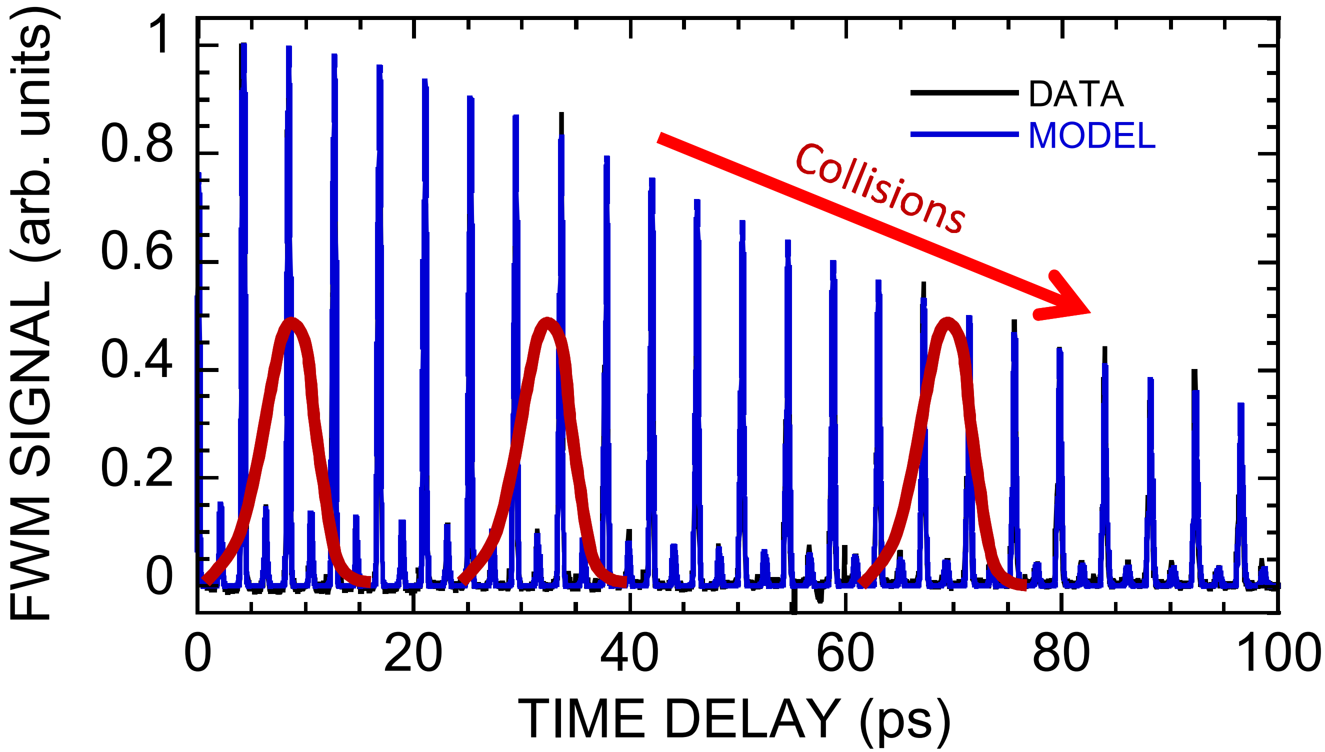


"*The story here is really in the time domain*"

Steady State Raman Spectrum in N_2

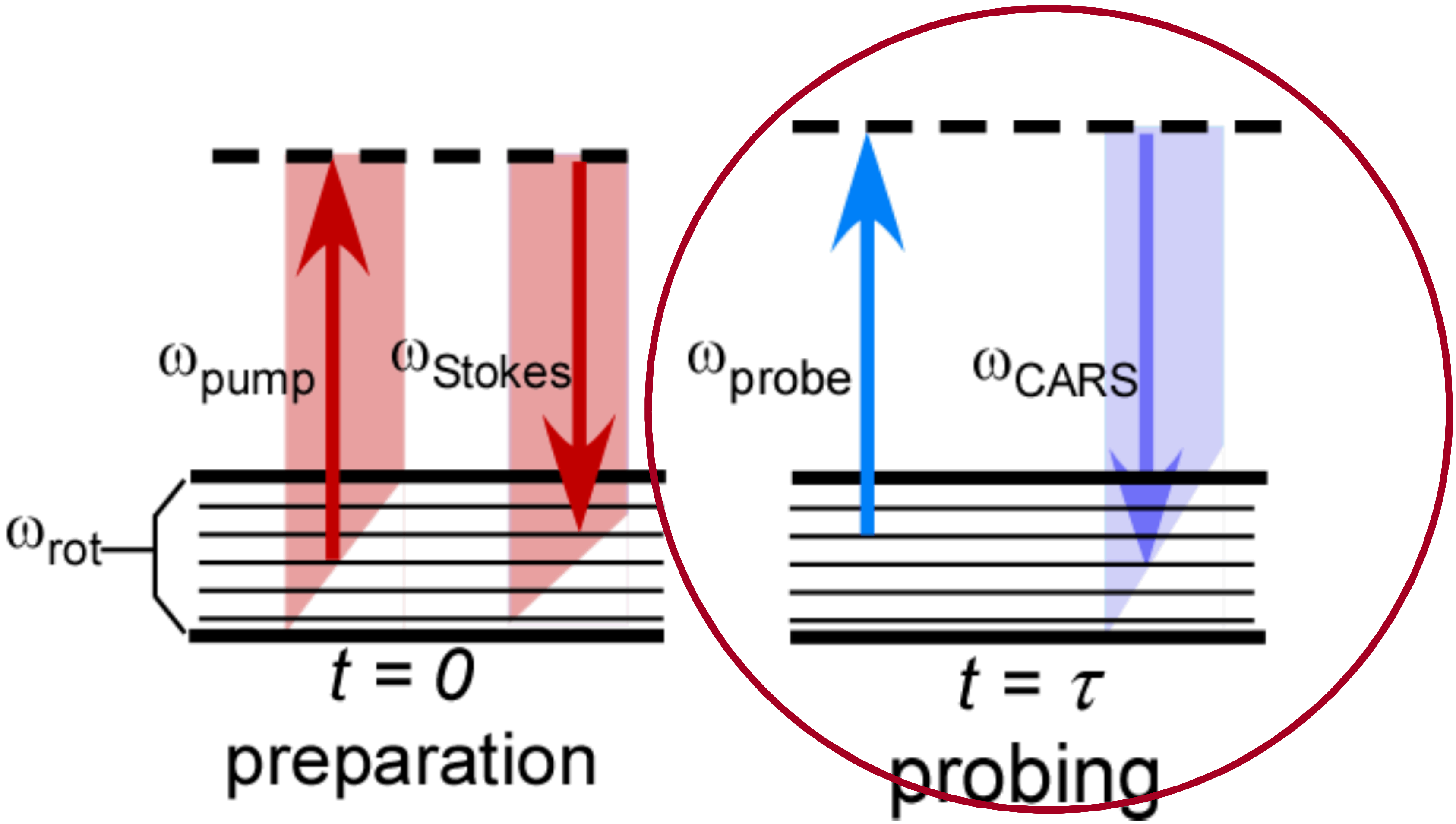


Time-domain Raman response in N_2



$$\chi = \sum_J W_J \exp\left(i\omega_J t\right) \exp\left(-\Gamma_J t\right)$$

Probe Step



Probe Step and Spectral Synthesis

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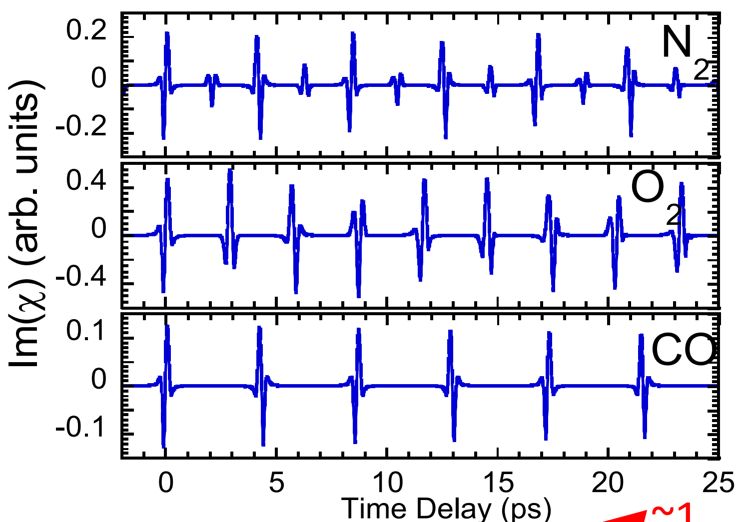
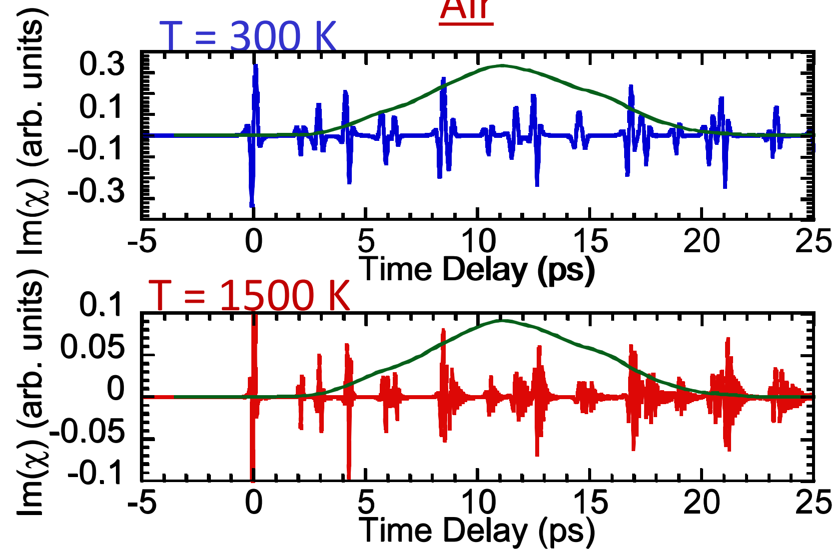
Pure Gases, T = 300 K



~1

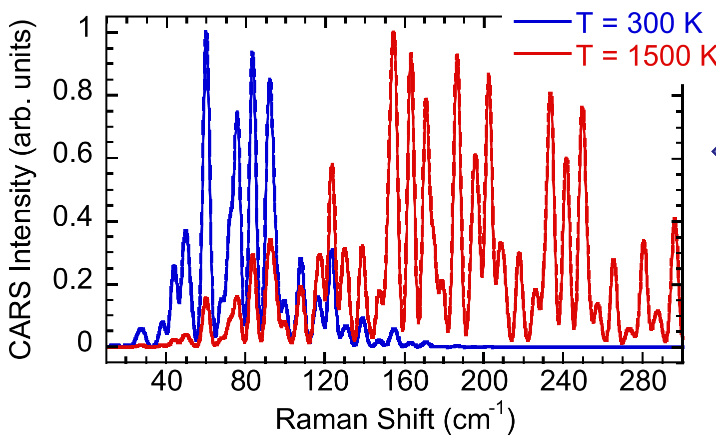
$$\chi = \sum_J W_J \exp\left(i\,\omega_J\,t\right)\exp\left(-\Gamma_J\,t\right)$$

Air



~1

$$\chi = \sum_k X_k \sum_J W_J^{(k)} \exp\left(i\,\omega_J^{(k)}\,t\right)\exp\left(-\Gamma_J^{(k)}\,t\right)$$



FFT

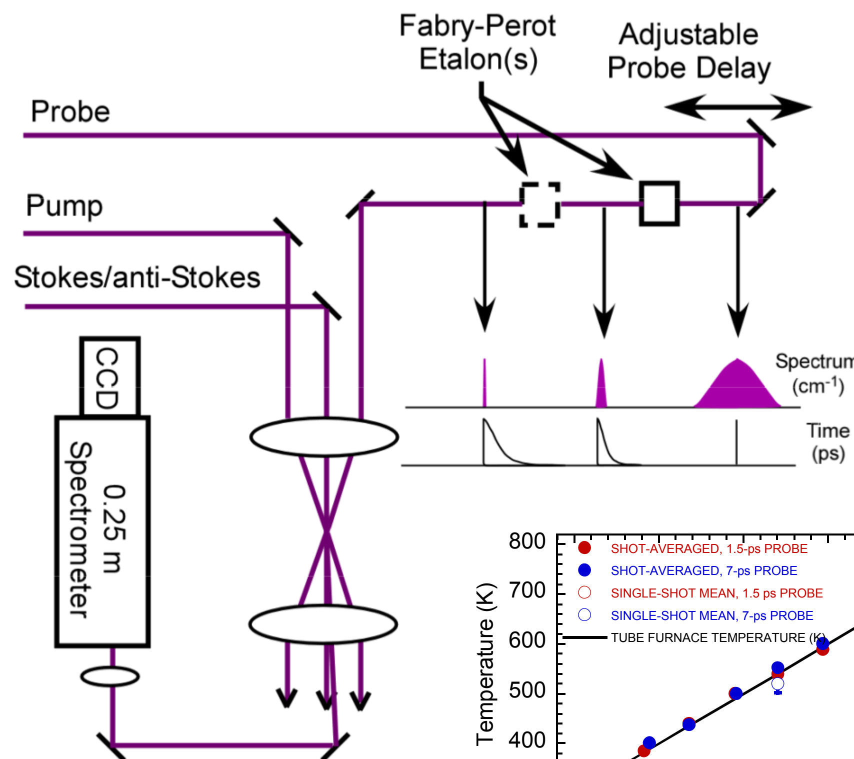
$$E_{CARS}\left(t\right) = E_{pr}\left(t-\tau\right)\times\chi\left(t\right)$$

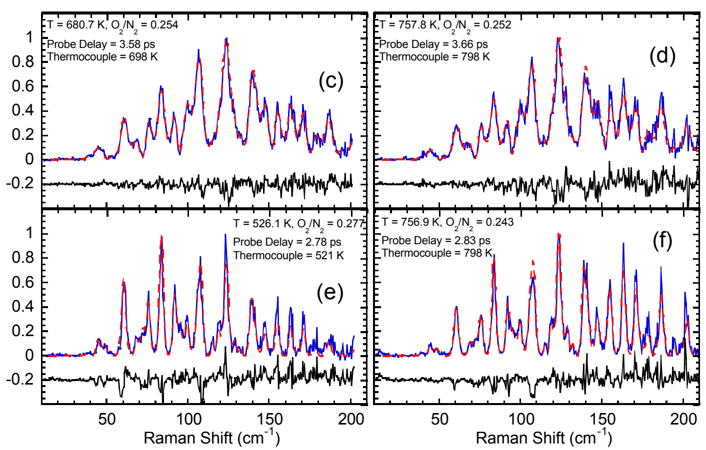
Initial Measurements via “Bandwidth Carving”

- Use filter to remove probe bandwidth
- Broadens pulse to ps regime

$$\Delta t \, \Delta\omega \leq const.$$

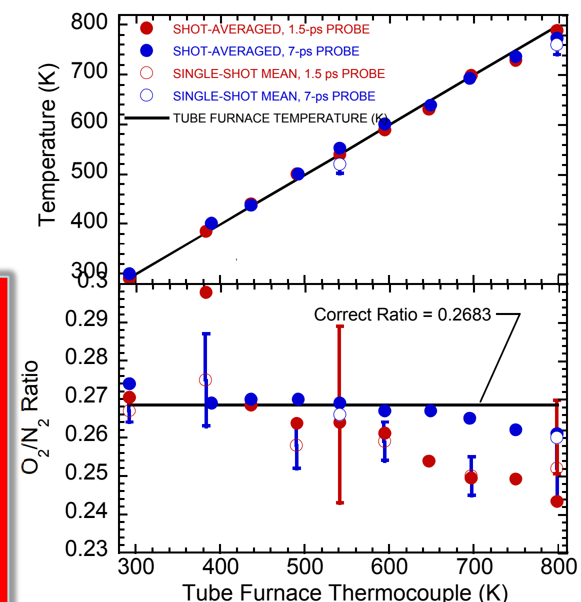
- Very inefficient (0.8 to 2.4% or less transmission)
- Good accuracy
- Outstanding precision
- Only applicable up to T ~ 800 K





Single-laser-shot spectra

Must generate higher probe-pulse energies!



S.P. Kearney *et al.*, *Optics Express* **21** (2013)

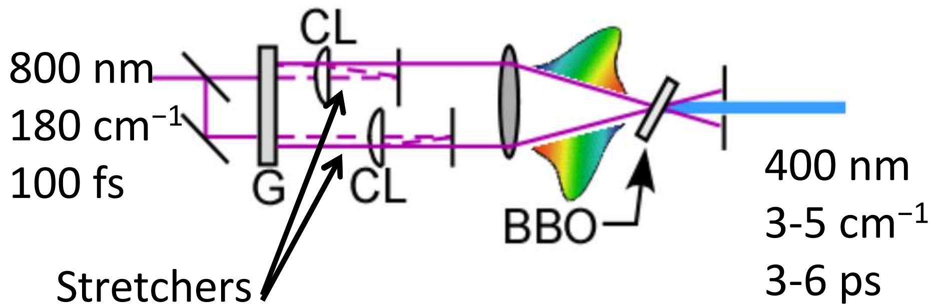
Second-Harmonic Bandwidth Compression (SHBC)

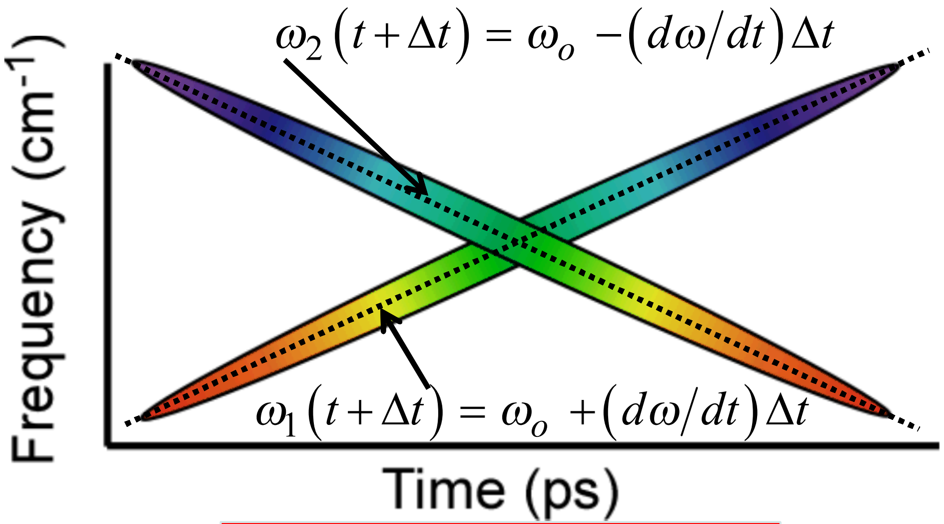
- Commercial device (Light Conversion)
- Converts fs radiation at 800 nm to ps radiation at 400 nm
- Grating pulse stretchers
- **Phase-conjugate** temporal chirps imparted upon broadband fs pumps
- Sum-frequency generation in BBO
- Output linewidth 3.5-4.0 cm^{-1}

$$\Delta\omega_{sfg} \sim d\omega/dt$$

$$\Delta\omega_{sfg} \sim (\Delta t)^{-1}$$

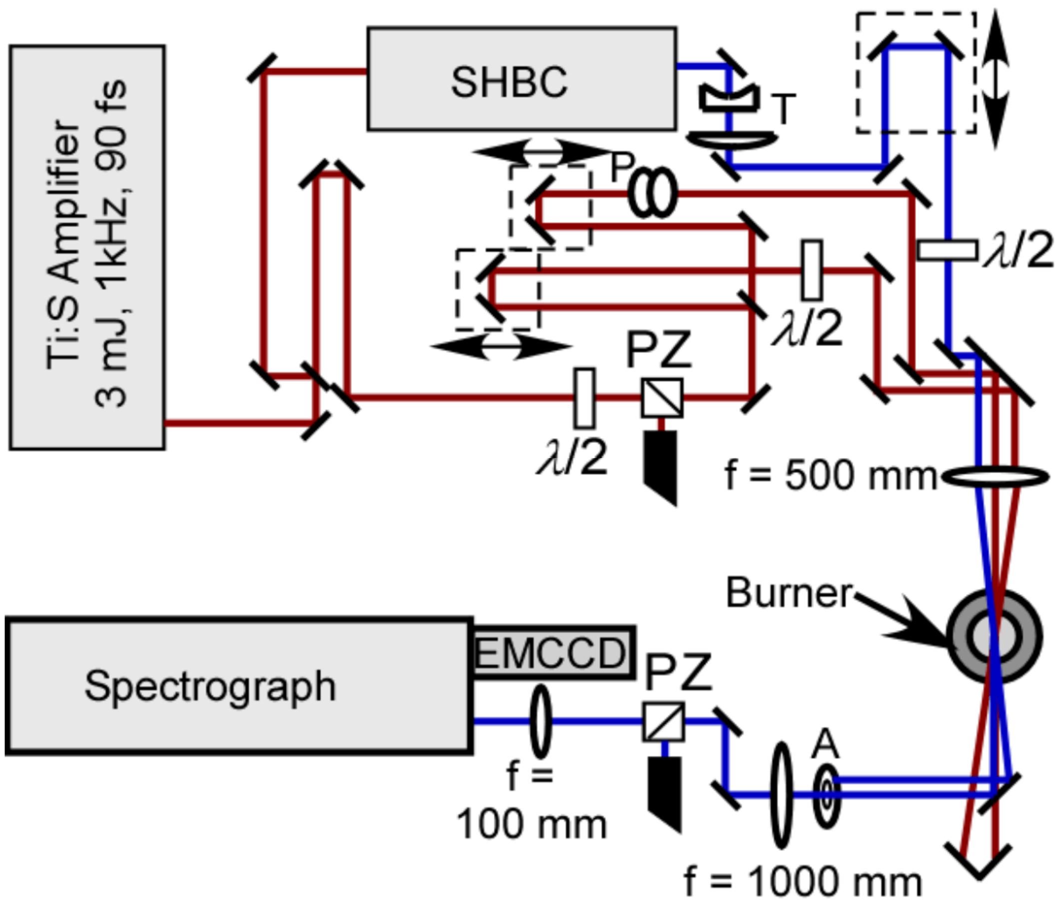
- Conversion efficiency: 35-50%!
- Output pulse energy: 1-1.4 mJ!





$$\omega_{sfg} = \omega_1 + \omega_2 = 2\omega_o$$

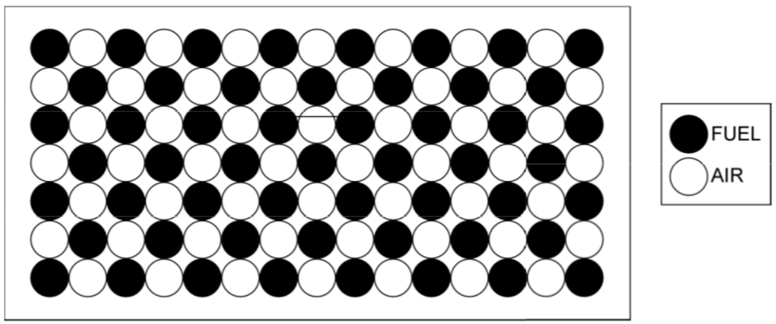
fs/ps SHBC CARS Instrument

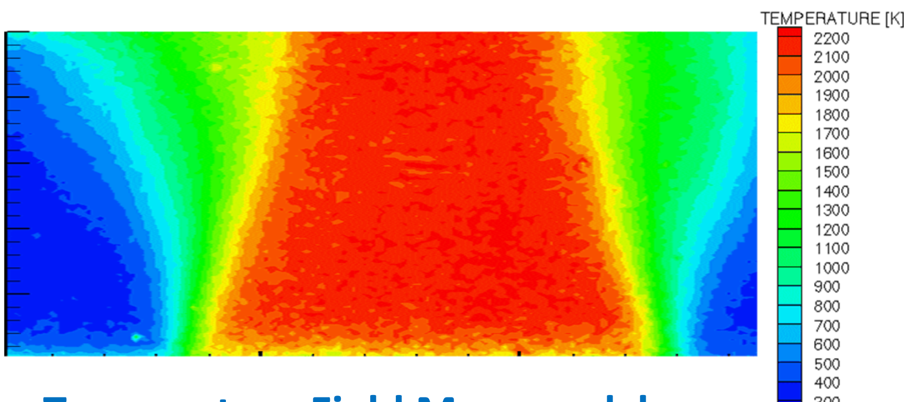


Assessment in Canonical Flat Flames

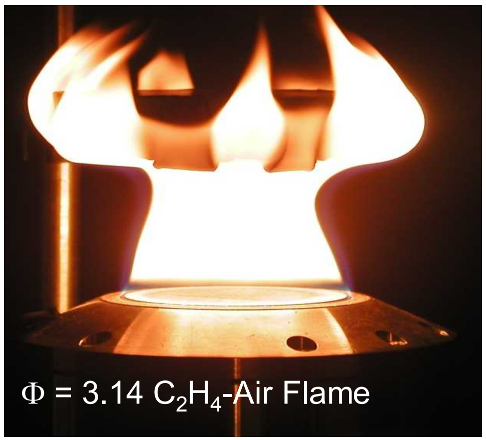
- ***Hencken Burner: Hydrogen/air flames***
  - Compare to adiabatic equilibrium
  - “Simple” fuel
  - Wide range of temperatures/products

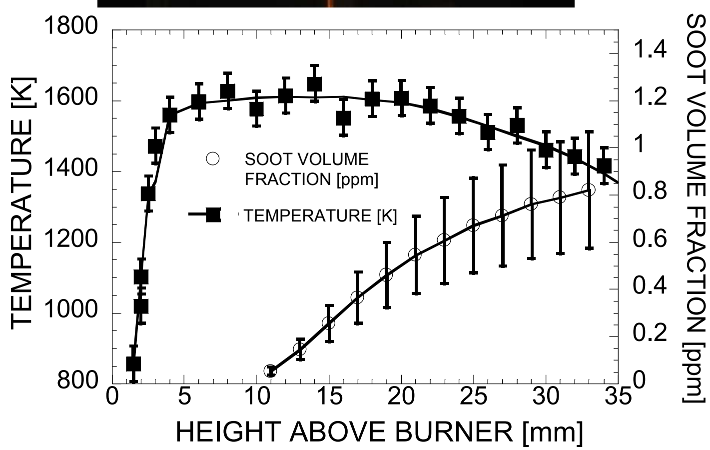
- ***McKenna Burner: Ethylene/air flames***
  - Compare to existing ns-CARS results
  - Hydrocarbon fuel with soot
  - Benchmark flame for soot & LII





Temperature Field Measured by Rayleigh Scattering (CH_4/air)



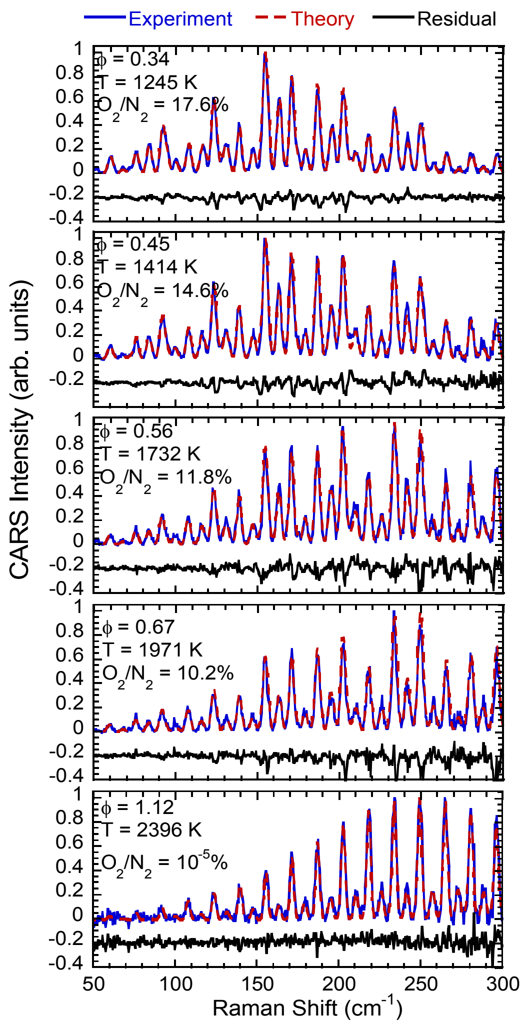


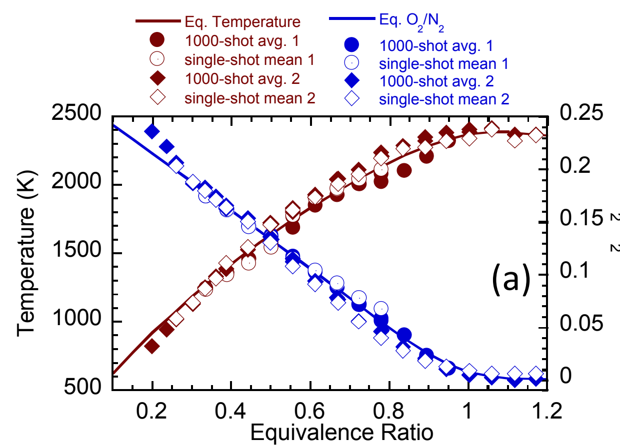
Results from H_2/air Hencken flames

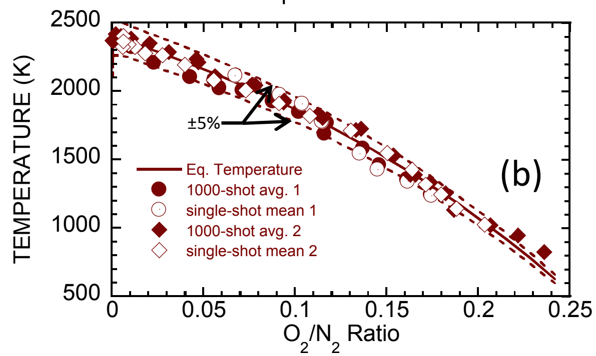
Sandia National Laboratories

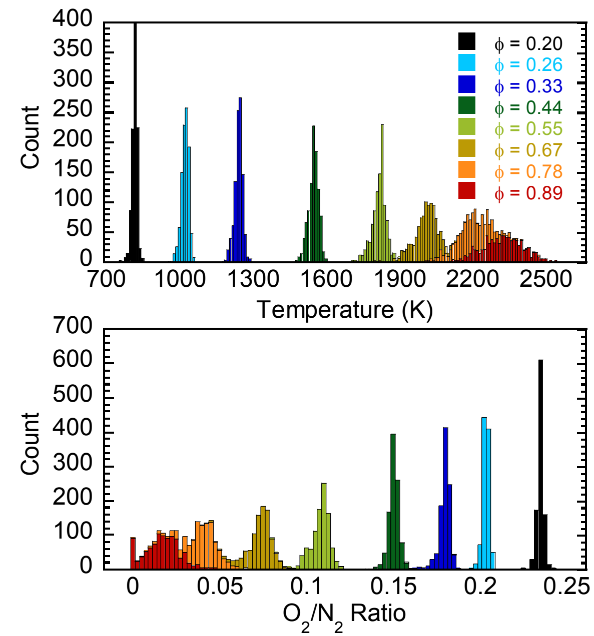
- ***Three orders of magnitude signal enhancement w/ SHBC***
- ***Single-shot measurements up to T = 2400 K***
- ***Temperature/O_2 measurements within 5% of equilibrium***
- ***Precision is outstanding at 1-2% for sufficient SNR***

Single-Shot at 1 kHz



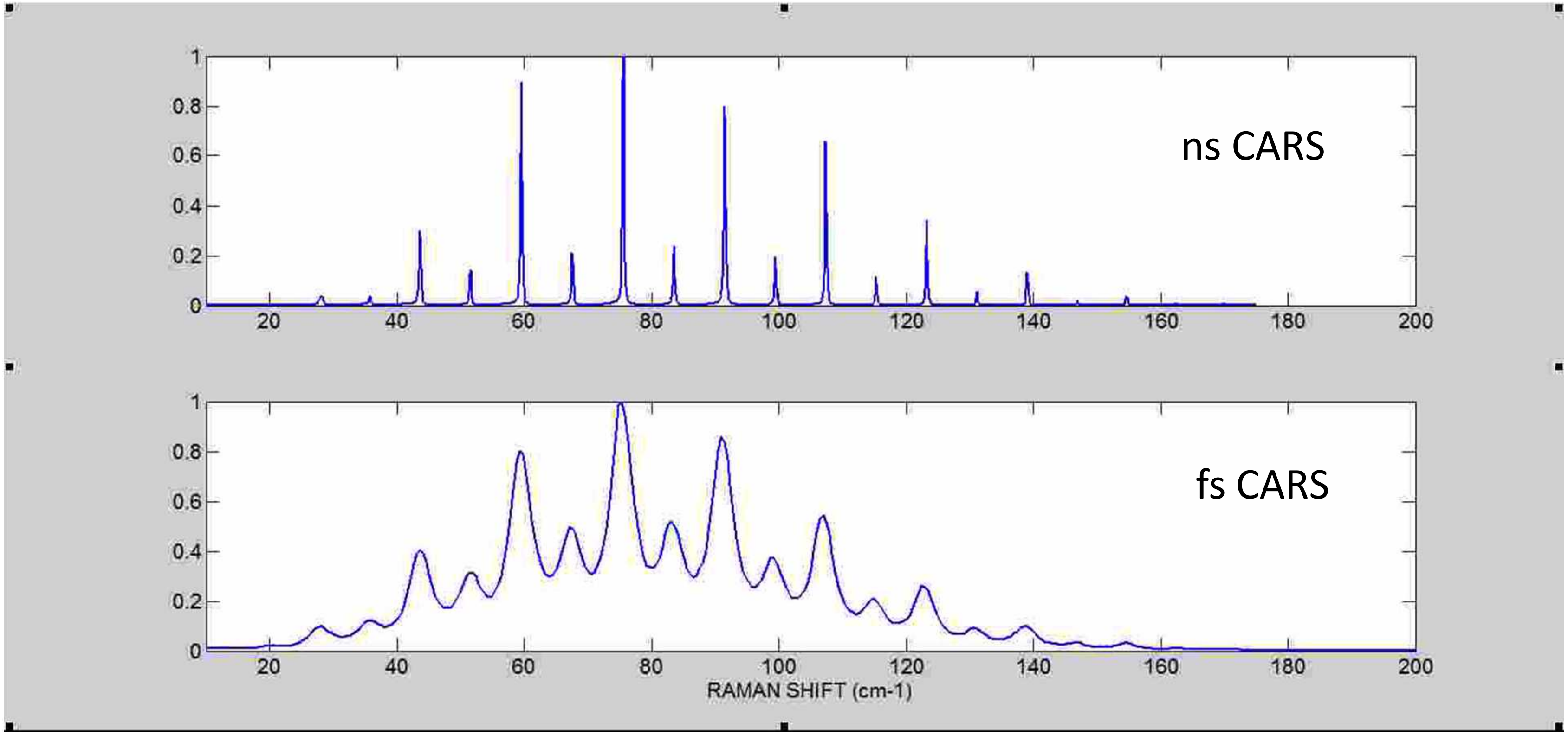






Kearney, *Combust. Flame* **162** (2015).

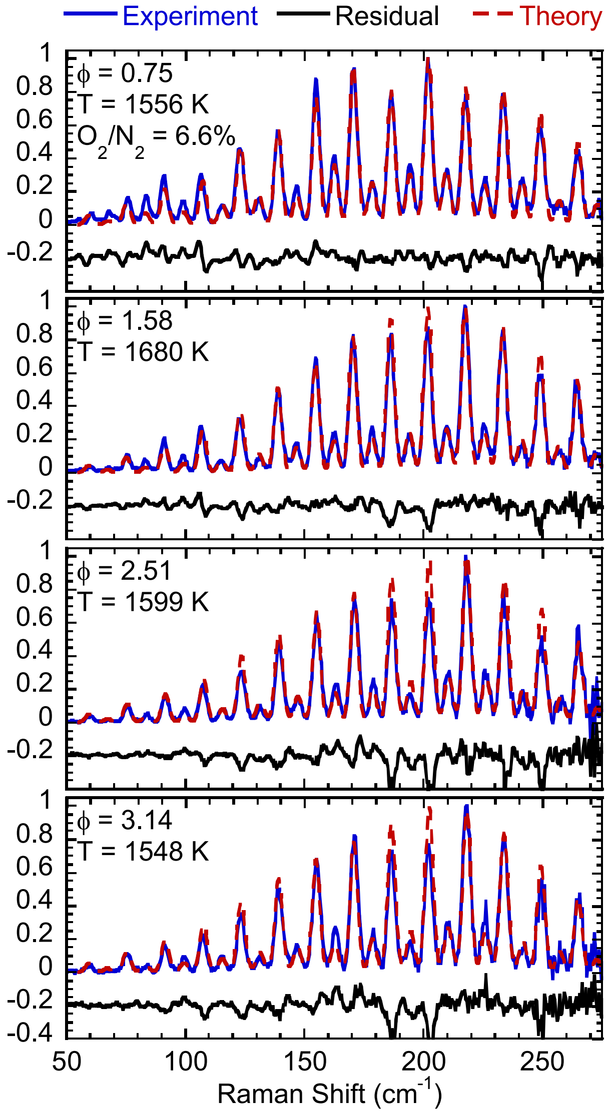
Low noise fs preparation pulses can result in higher single-laser-shot precision



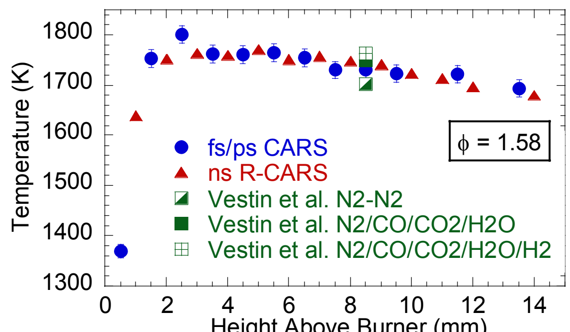
Room-temperature N_2 spectra

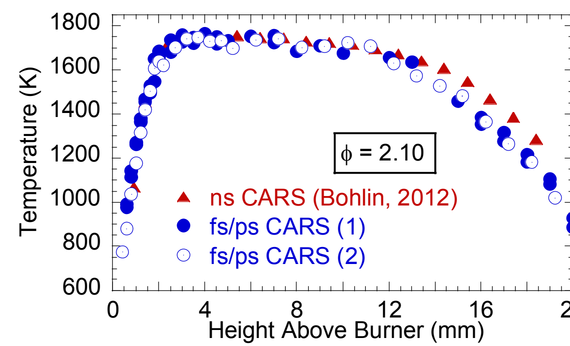
C_2H_4/air McKenna flame results

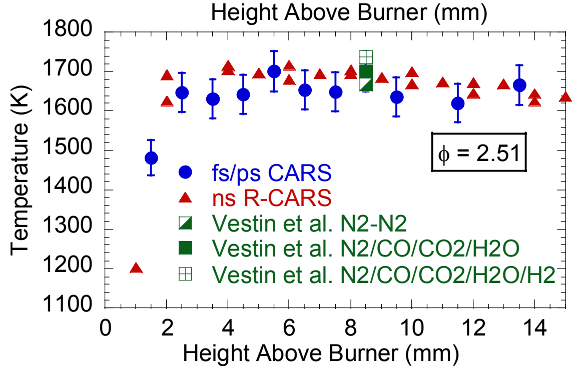
Single-Shot at 1 kHz

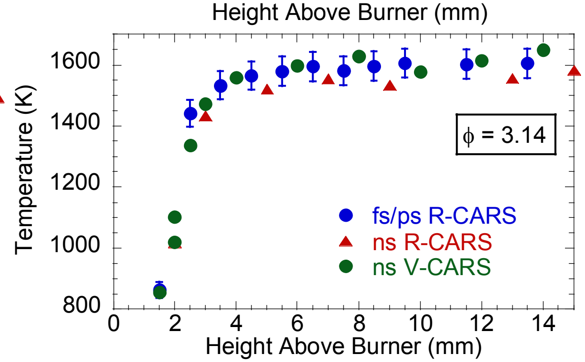


- ***Spectra acquired for fuel-lean to rich sooting flames***
- ***High-quality fits observed for ϕ < 1***
- ***Systematic bias toward "underfit" of isolated lines for fuel-rich flames***
- ***Fitted temperature appears to be robust***
- ***Reliable spectra obtained in <u>sooting regions</u> of the flame***







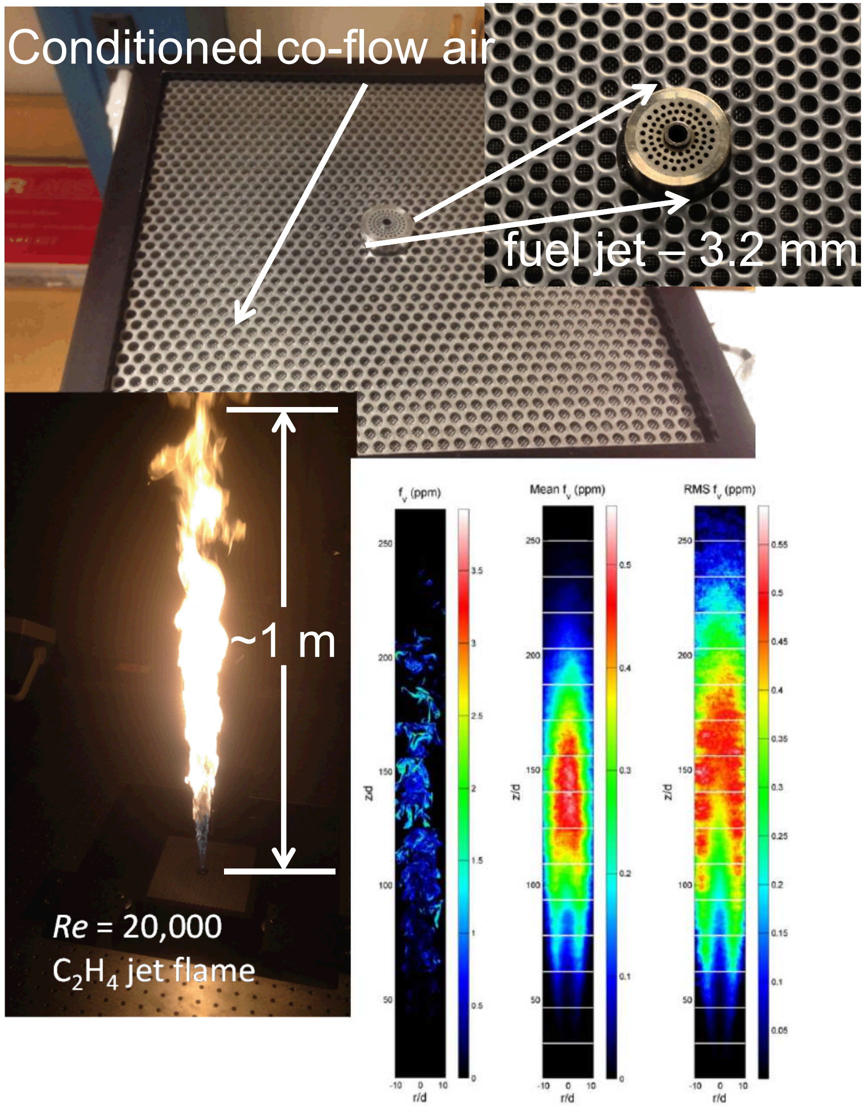


Applications

Turbulent ethylene jet flame burner

Sandia National Laboratories

- *Sandia CRF design (Shaddix)*
- ***Pilot-stabilized canonical turbulent jet flame is ideal for model development***
- *Ongoing measurements*
  - *Temperature/O2 by fs/ps rotational CARS at 1 kHz*
  - *Soot LII at 10 Hz*
- ***Soot/smoke yield can be varied by vitiation, dilution, or fuel type***
- *Baseline case → pure C_2H_4 fuel*
- ***Provides fully turbulent temperature/soot profiles***
- *Mean soot fv ~ 0.6 ppm, jet-fuel pool fire is ~ 1 ppm*



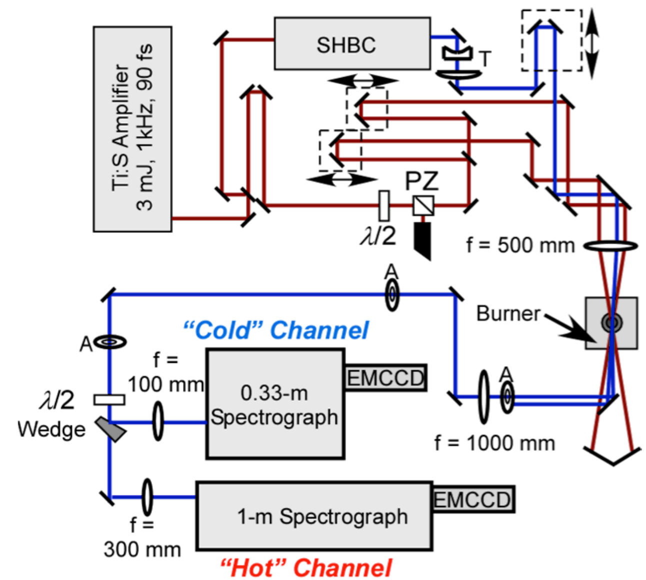
Soot LII data (Shaddix, 8300)

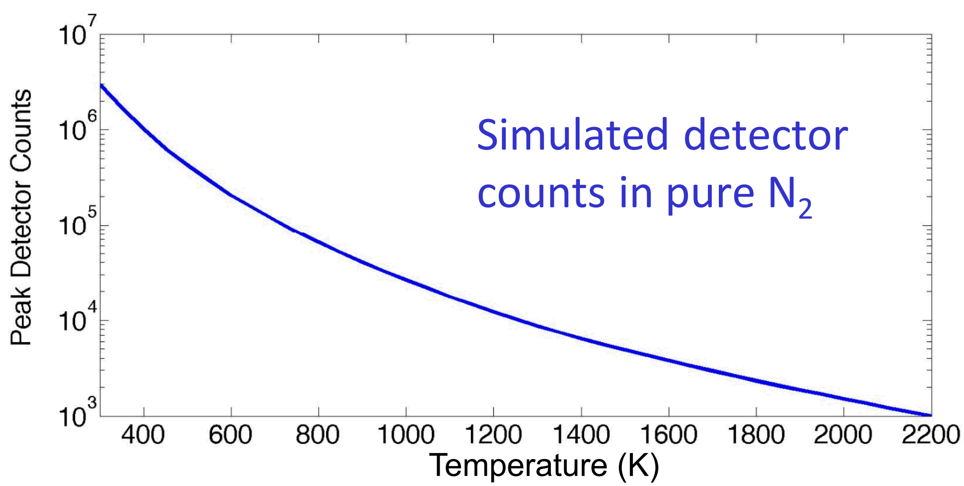
Movie o e

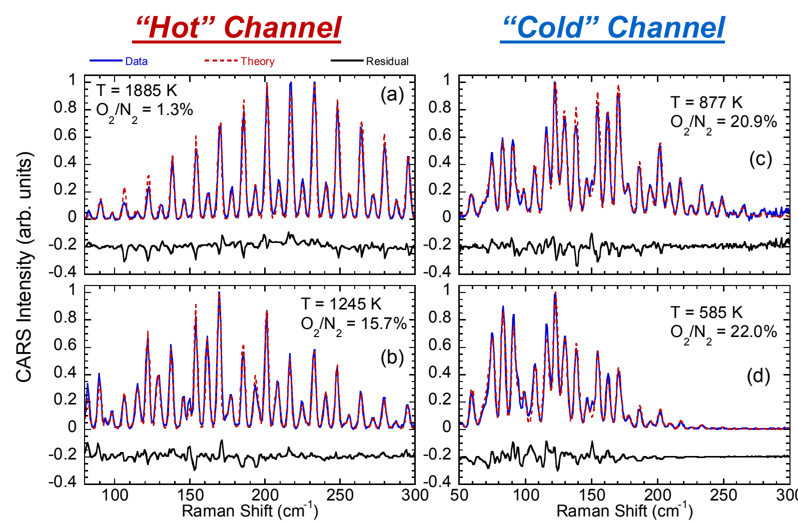
Sandia National Laboratories

Two-channels for enhanced dynamic range

- ***CARS signal strength varies by ~3 orders of magnitude between room and flame temperatures***
- ***The highly fluctuating temperatures in turbulent flames make dynamic range a challenge***
- ***Two-channel detection system implemented***



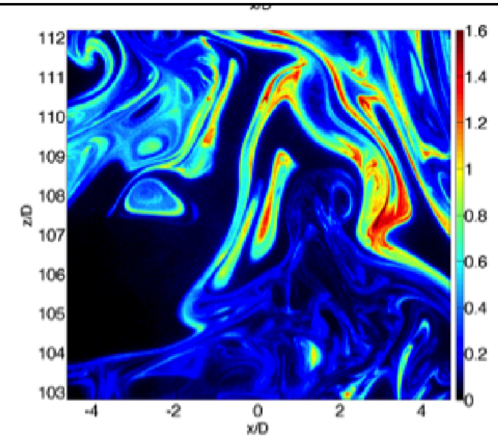


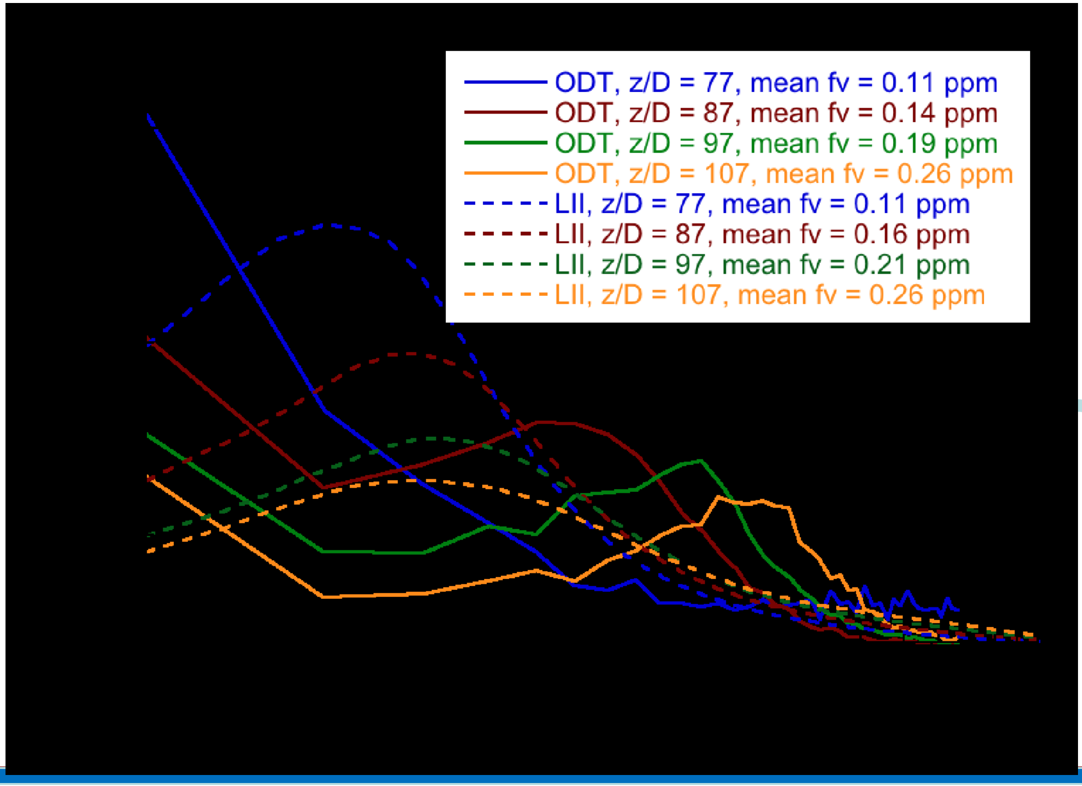


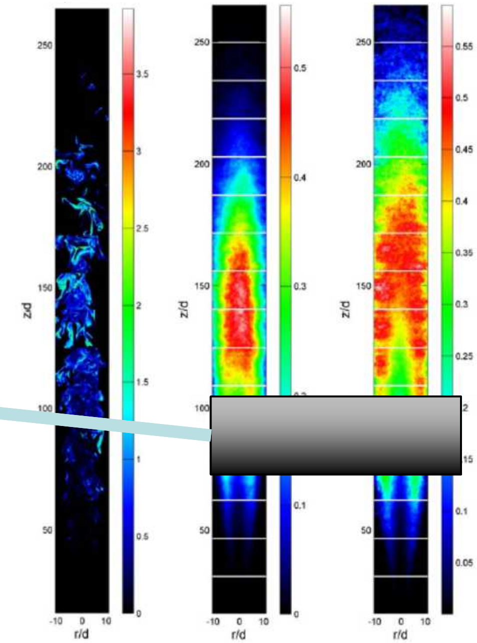
Single-Laser Shot Spectra

LII soot measurements from turbulent jet flame

- *Single-shot LII image data for z/D ~ 75-100*
- *500 laser shots in 29 mm (9.1 D) field of view at each height*
- *Soot pdf data estimated within field of view at each height*
- *Mean and rms soot values consistent with Shaddix data*
- *One-dimensional turbulence simulations tuned to match mean soot f_v*



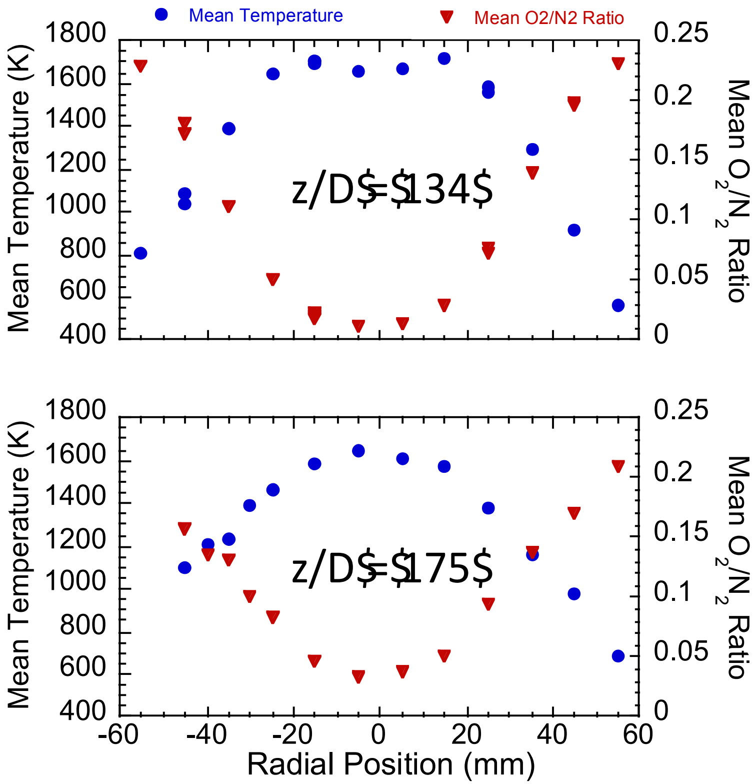


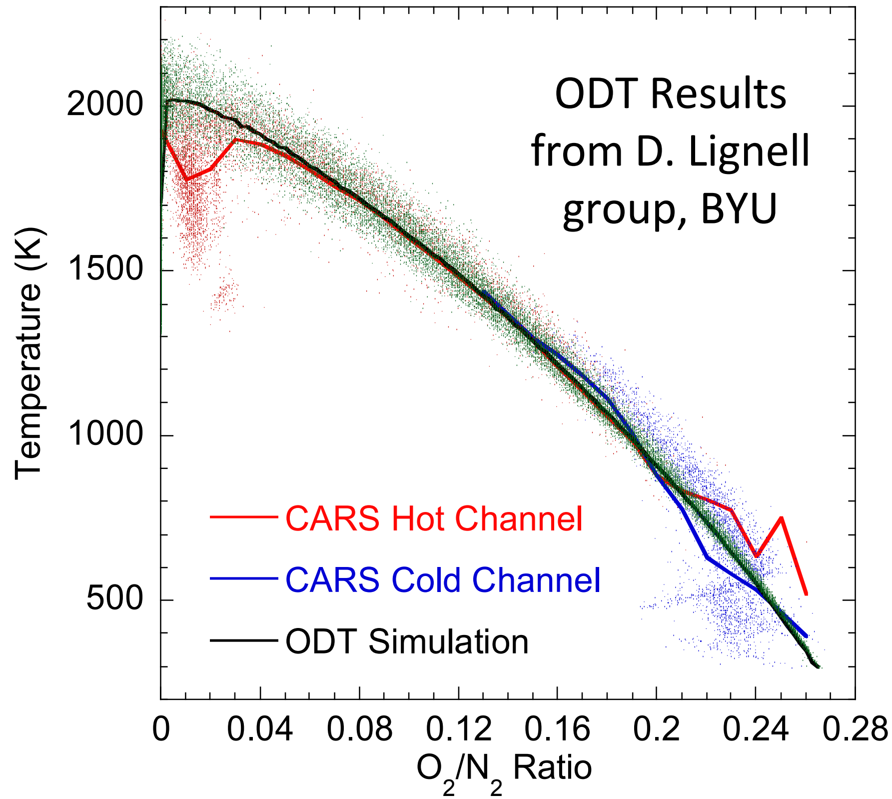


Soot LII data (Shaddix, 8300)

Temperature and O_2 Measurements

Sandia National Laboratories





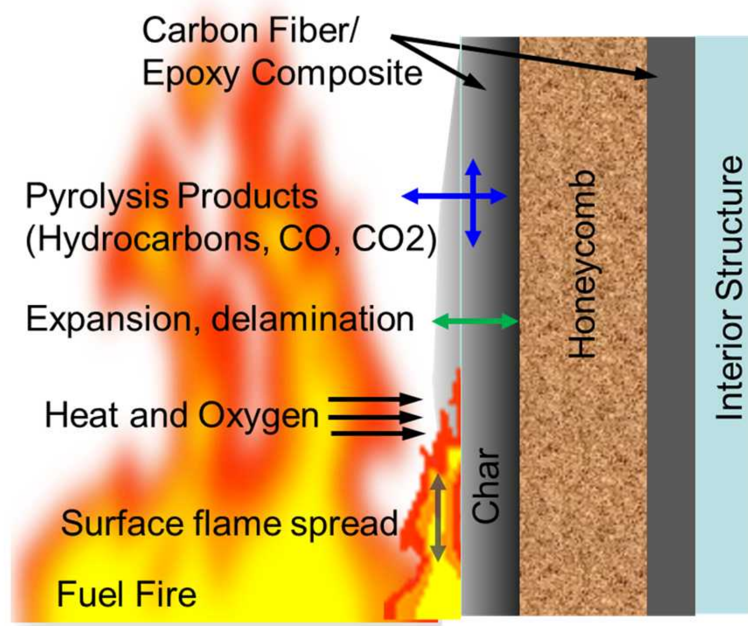
- ***Results obtained in region of peak soot loading, mean f_v ~ 0.5 ppm***
- ***Valid, detectable signal on 95-99% of all laser shots***
- ***ODT simulations predict scalar response when soot source in calibrated to LII***

Carbon-Epoxy Solid Fuels

"Crashed B2" Federal Aviation Administration Photo

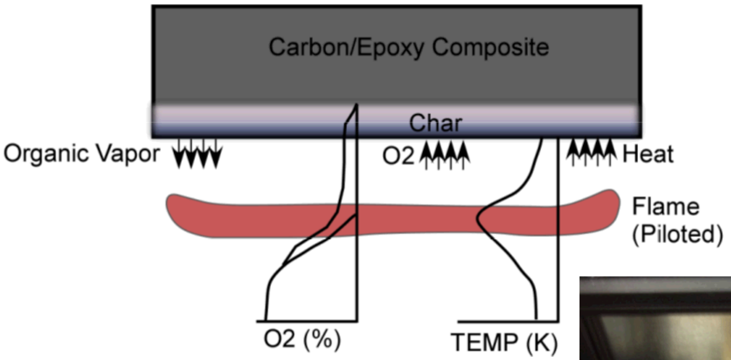
- *Composite fires are not well characterized*
    - *Aviation fuel fire heats composite*
    - *Epoxy thermal decomposition*
    - *Heat and mass is transported to interior*
    - *Char formation*
    - *Oxidation reaction of carbon fibers?*

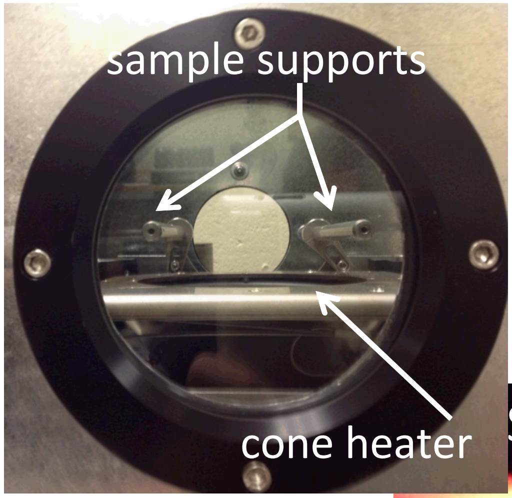
- *Carbon-epoxy composite materials are being used in a wide variety of military and civilian aircraft*
- *Accident scenario presents new risks in addition to hydrocarbon burn problem*
- *Fundamental data are lacking*
- *Surface Oxygen transport?*
- *Temperature profiles*

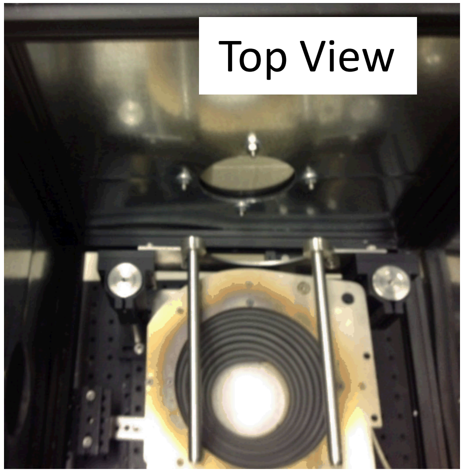


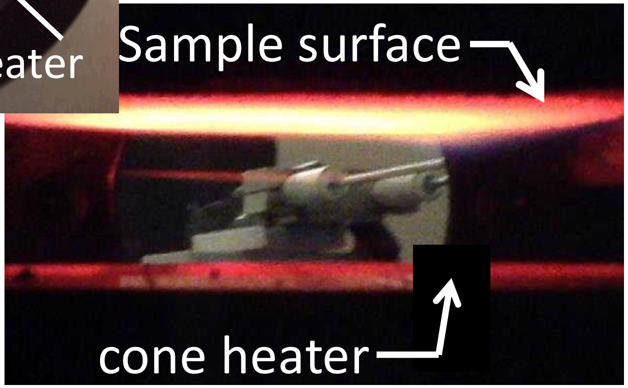
Controlled Carbon-Epoxy Burn Experiment

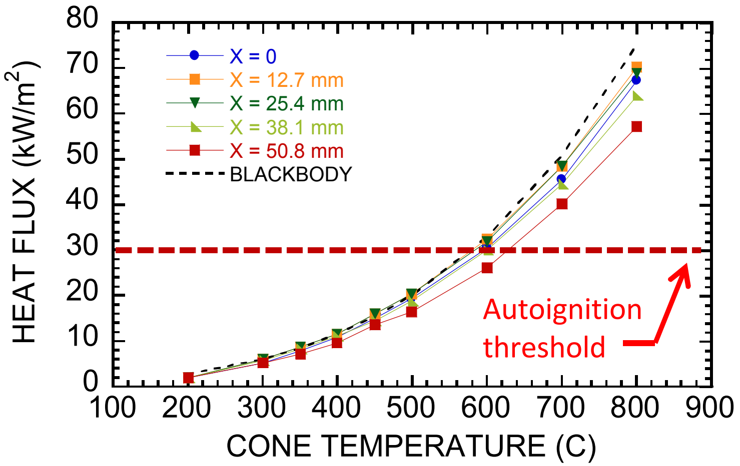
- *100-mm x 150-mm composite samples*
- *3-mm thickness*
- *Backside insulated*
- *Suspended over "cone heater"*
- *Uniform radiative heat flux over 100-mm dia area*
- *CARS T and O_2 data recorded on bottom side of sample*









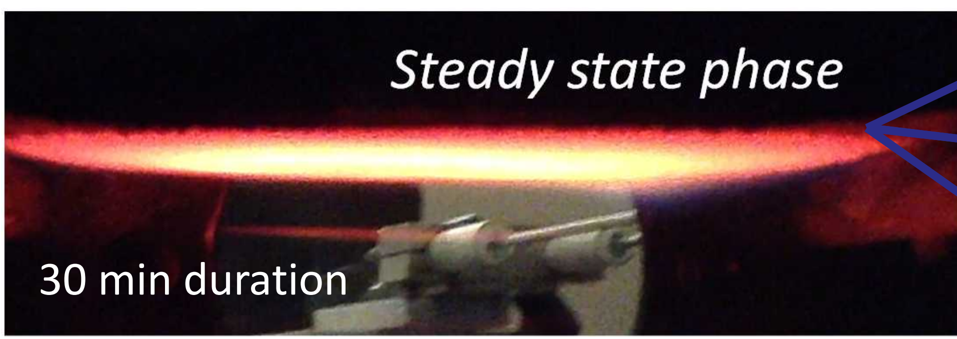


Cone Heater Flux

Observed sample decomposition/burning behavior

- Autoignition at heater temperature ~600 °C or q″ = 31 kW/m^2
- Combustion of ***epoxy binder*** material
- ***Heavily sooting***—absorbs almost all laser light near surface

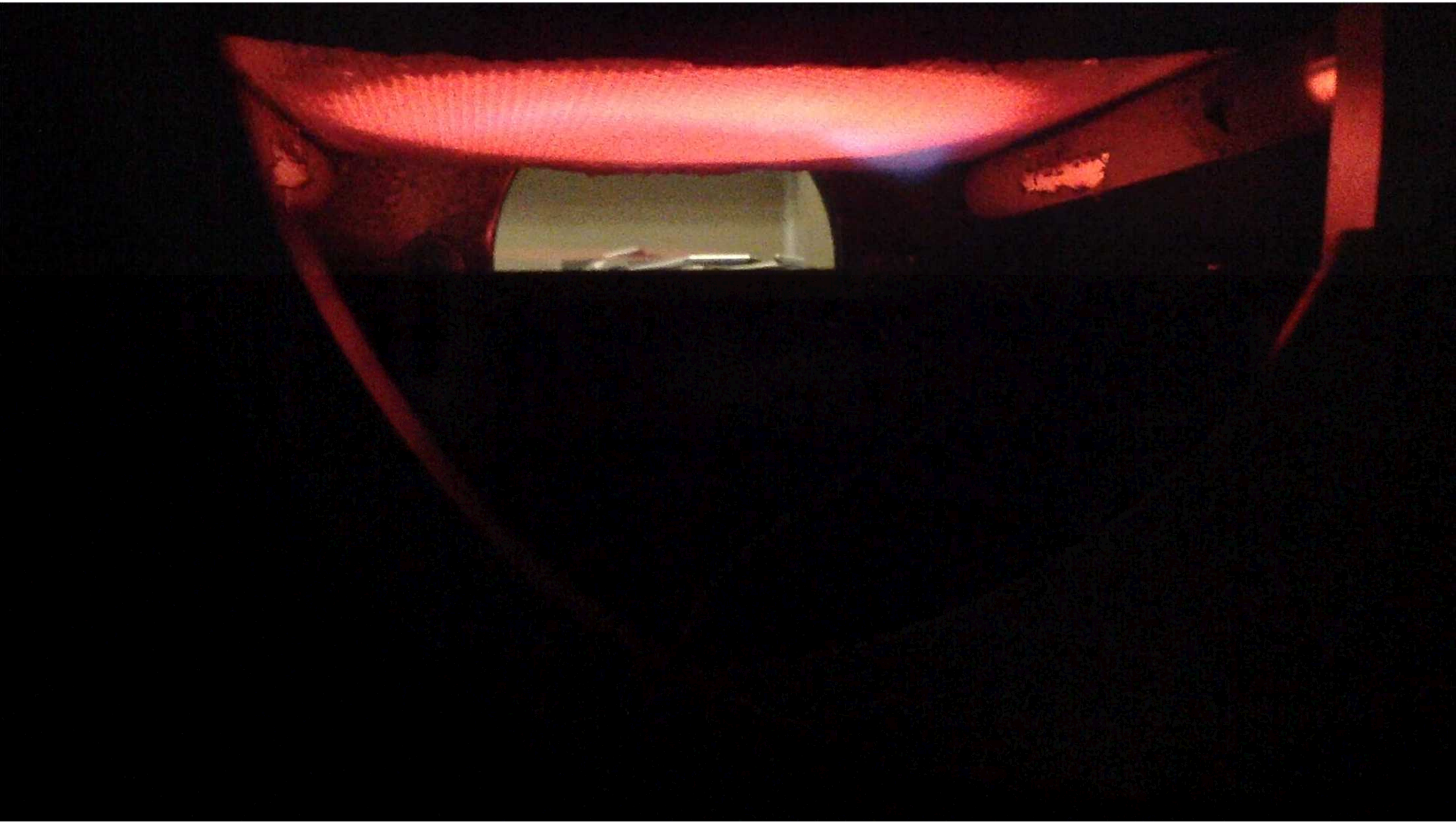
- Transitional phase
- Binder nearly consumed
- Pockets of sooty flame meander about surface



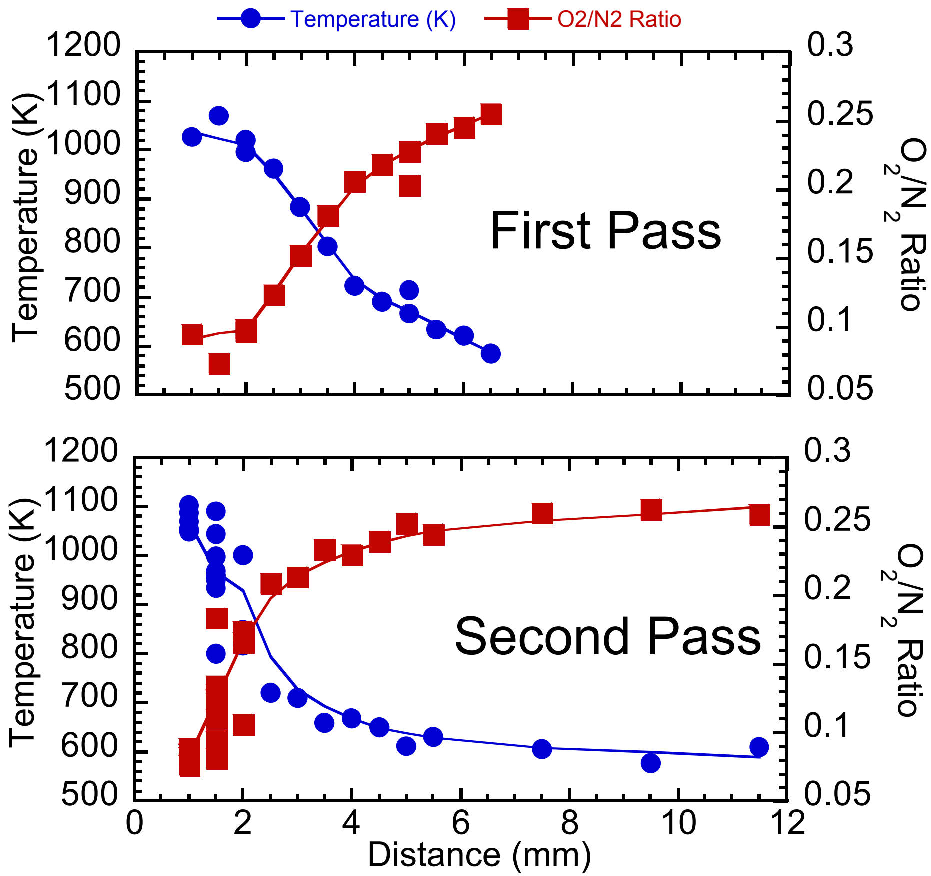
- Consumption of carbon fibers
- Perhaps some char consumption
- Both blue and yellow/orange flame emission

Ignition and sooting epoxy burn

“Steady state” fiber consumption

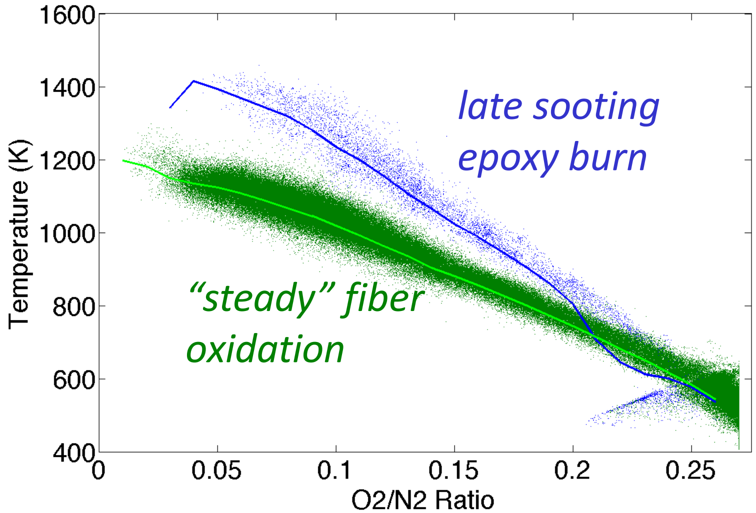


Temperature/O_2 Profiles: ***Point Measurements***



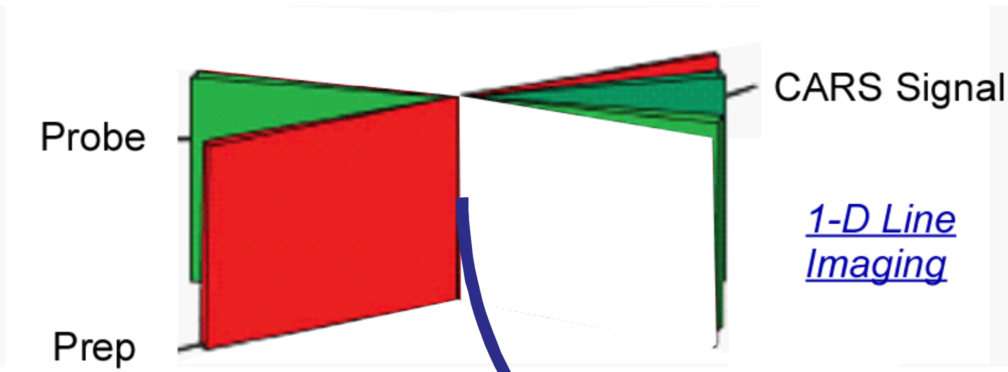
"Near-wall" temperature/oxygen profiles

- Sampled for 10 seconds (10,000 laser shots) at each point.
- Sample/chimney/heater translated vertically
- Constant surface recession during measurements
- 2.5 mm shift of second profile

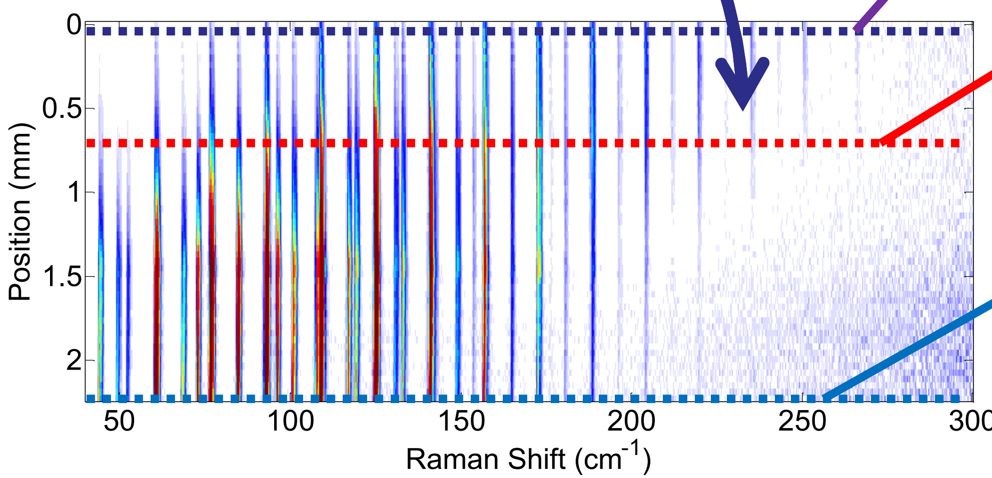


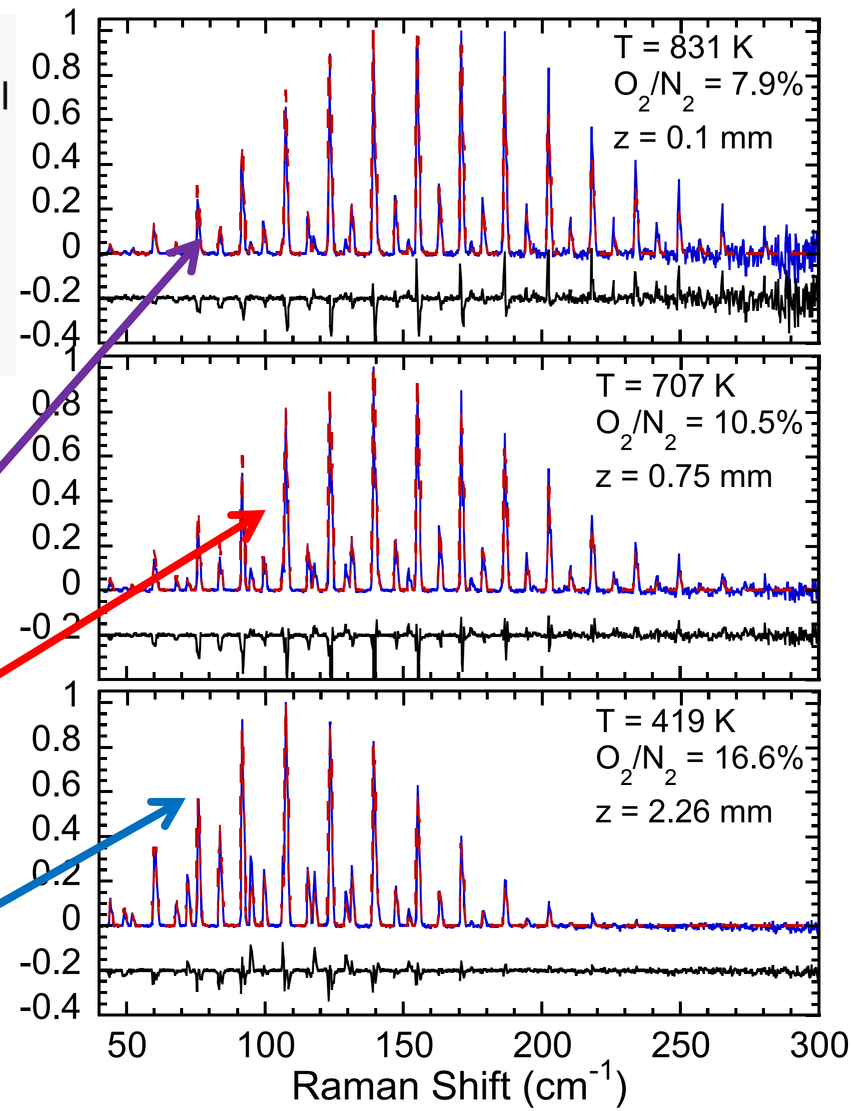
Temperature/oxygen correlation

1-D CARS Line Imaging at Sandia/CA



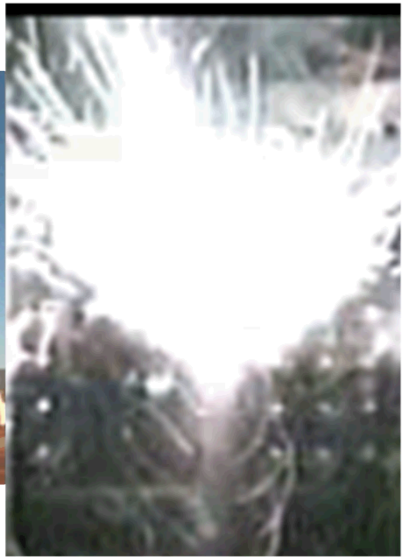
Intersection of light sheets imaged onto spectrometer slit/CCD

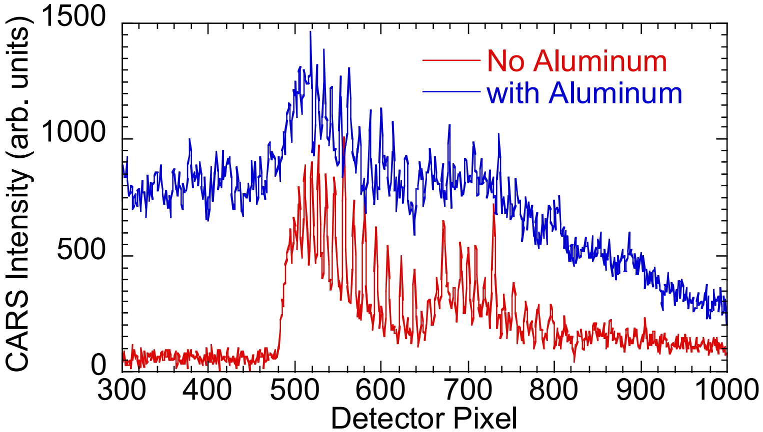




Metallized Propellant Fires

- *Metal-burn problems are important to the safety and performance of a number of DOE/DoD systems*
  - *Missile-platform safety*
  - *Igniters*
  - *Energetic materials*
- ***Metal burn represents an extraordinarily hostile environment for laser diagnostics***
  - ***Very high temperatures – T > 3000 K!***
  - ***High luminosity***
  - ***Scattering***
- ***Previous gas-temperature measurements with ns CARS were unsuccessful***
  - ***Metal-particle-induced breakdown/plasma formation***
  - ***Disastrously high levels of nonresonant background***

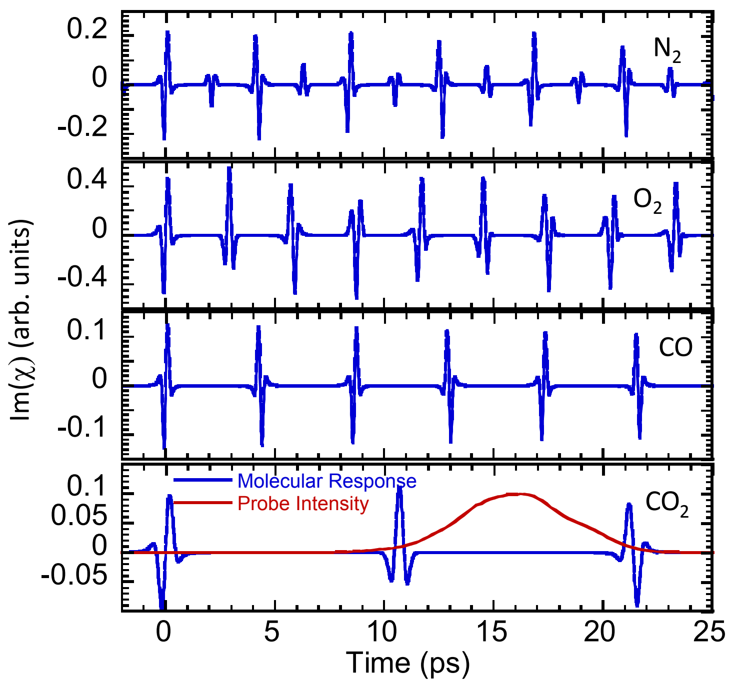


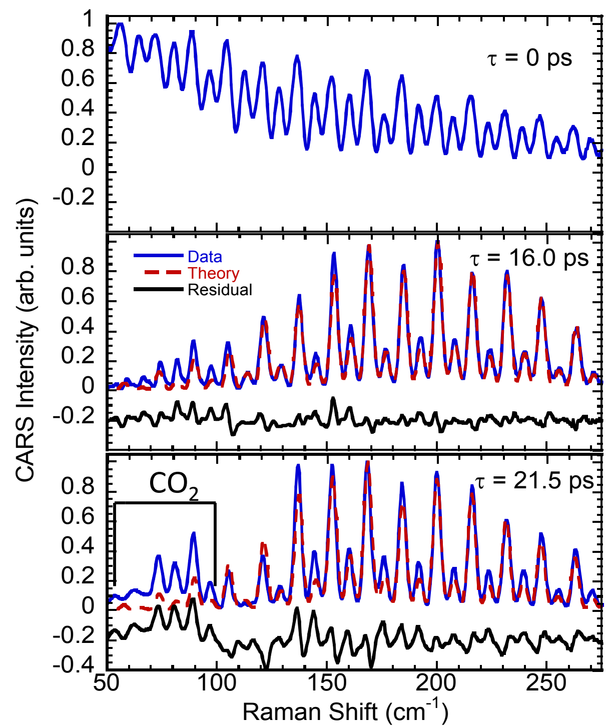


ns-CARS spectra with and without Al particles

Advantages of ultrafast CARS for metal-burn problems

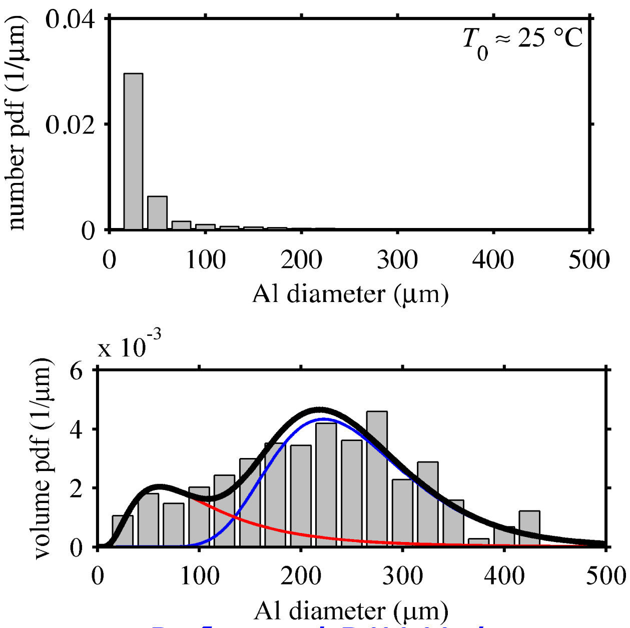
- ***Low total pulse energy***
  - ***~ 1 mJ or less***
  - ***Reduces the likelihood of breakdown-type interference***
  - ***Factor of ~40-100 lower than with ns laser pulses***
- ***Time-gate elimination of nonresonant background***
  - ***Nonresonant signal arises from response of electrons to fs forcing***
  - ***Fast decay (fs)***
  - ***Raman-resonant signal results from much slower, nuclear response of the molecule***

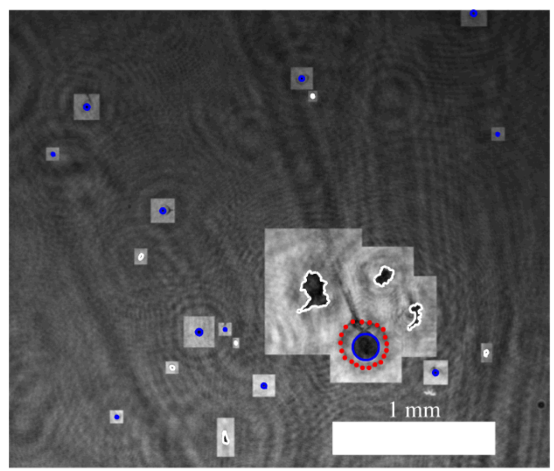




AP Propellant Strand Burner Experiments

- ***Proof-of-concept using existing rotational CARS instrument***
- ***Acquired ~60,000 single-laser-shot spectra over strand-burner duration***
- ***Hostile environment for diagnostics***
  - ***Very high temperatures – fluctuations over 3,000 K!***
  - ***Dense field of large molten-metal particles 10-100 µm***
- ***Significant number of spectra rejected, but over 20,000 were retained for valid temperature measurements***





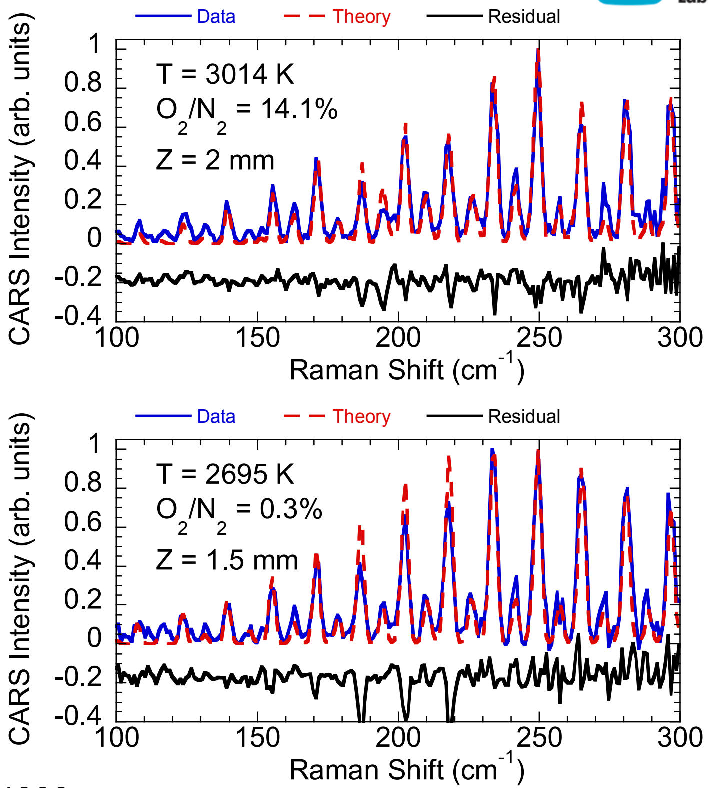
(b)

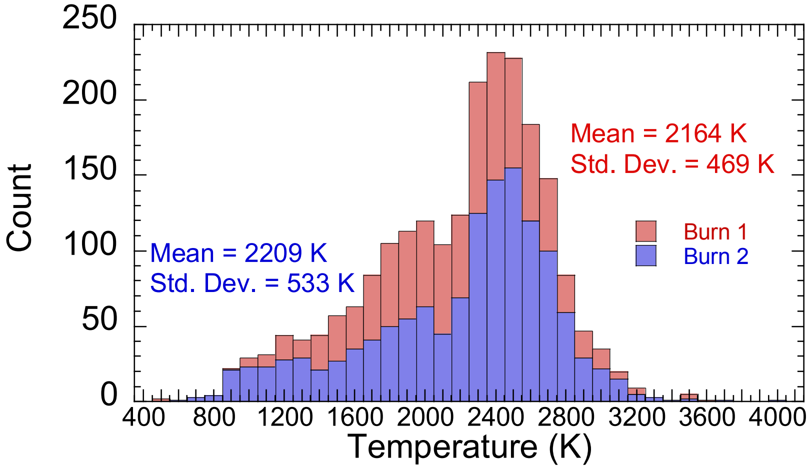
Refocued DIH Hologram and Al Particle Size Distributions (Guildenbecher, 1512)

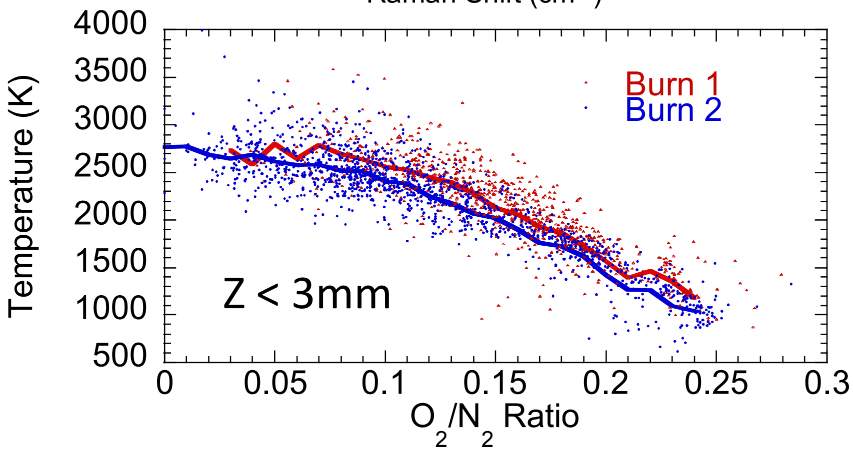
Video Recorded During CARS Measurements

CARS Results from Propellant Burns

- ***Results for CARS measurements within 3 mm of propellant surface shown***
- ***Height estimated from 1-kHz acquisition rate and burn rate of 800 μm/sec***
- ***Spectra dominated by N_2 and O_2 contributions***
- ***Cl2; HCl; NO; NO2 also present***
- ***Switch to vibrational CARS for follow-on experiments***
- ***Decrease measurement volume length***







Summary and Conclusions

- CARS is a powerful technique for temperature/species measurements in hostile and facility-scale environments
- Nanosecond V-CARS has been performed with LII in 2-m, sooting fire plumes
- We have cultivated femtosecond/picosecond R-CARS
  - SHBC implementation
  - Collision-free results with superior precision
- fs/ps R-CARS is being applied in challenging laboratory environments
  - Sooting, turbulent jet flames
  - Metallized propellants
  - Carbon-epoxy composites
- ***Whats next?***
  - Pressure measurements—monitor collisions on ps scales
  - Vibrational fs/ps measurements for propellants
  - Rocket motor plumes
  - 2-D CARS imaging w/ soot LII