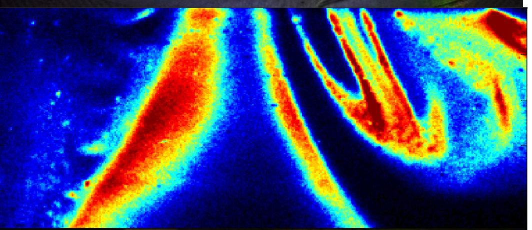
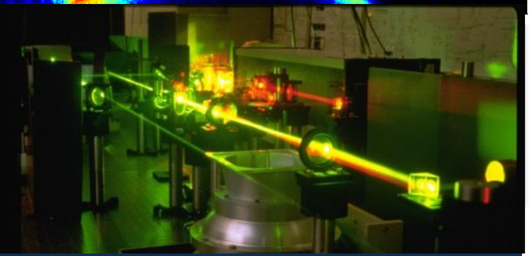
SAND2013-8970P

Laser Combustion Diagnostics for Sooting Flames and Turbulent Pool Fires





Sean Kearney

Engineering Sciences Center

Sandia National Laboratories

Albuquerque, NM 87185

(spkearn@sandia.gov)

Sandia National Laboratories

Exceptional service in the national interest

U.S. DEPARTMENT OF ENERGY

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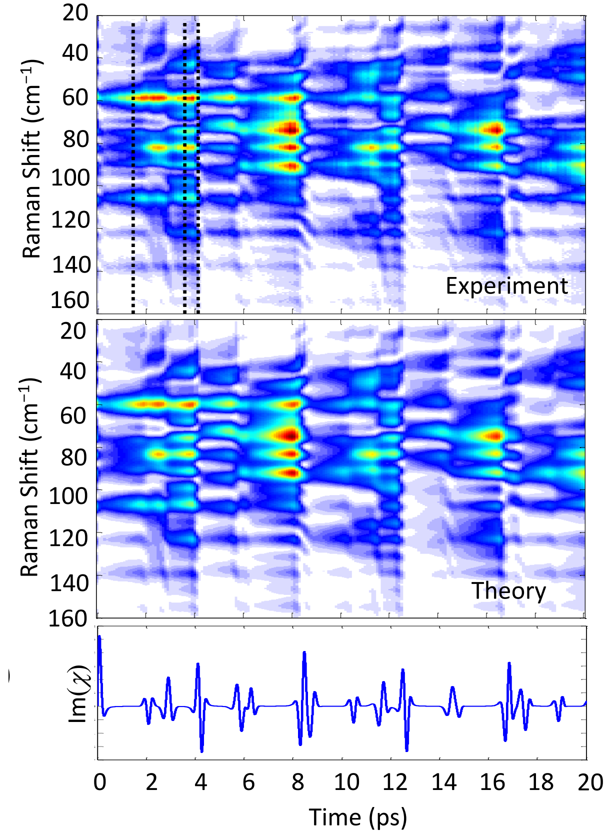
Overview

- Laser diagnostics in sooting and meter-scale fire systems (30 mins)
  - Sandia's motivation for fire research
  - Advantages of and challenges of using laser diagnostics for fire studies
  - Coherent anti-Stokes Raman scattering (CARS)
  - Laser-induced incandescence (LII)
  - Laboratory and field measurements
- Femtosecond CARS (15 mins)
  - Femtosecond shock interferometry (USI)
  - PIV measurements in unsteady wall turbulence

LII Diagnostic in a Full-Scale Fire Plume

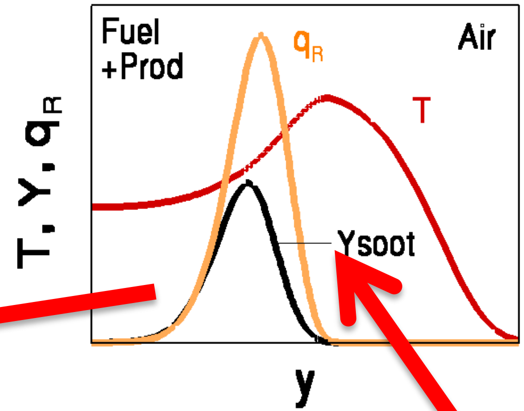


Femtosecond CARS

Radiative Heat Flux from Soot is the Dominant Source of Risk in a Fire-Accident Scenario

- Thermal radiation heat flux is generated and absorbed *locally* within the individual flame zones.
- Radiative transfer equation

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



Local soot & temp fields add up to global heat flux

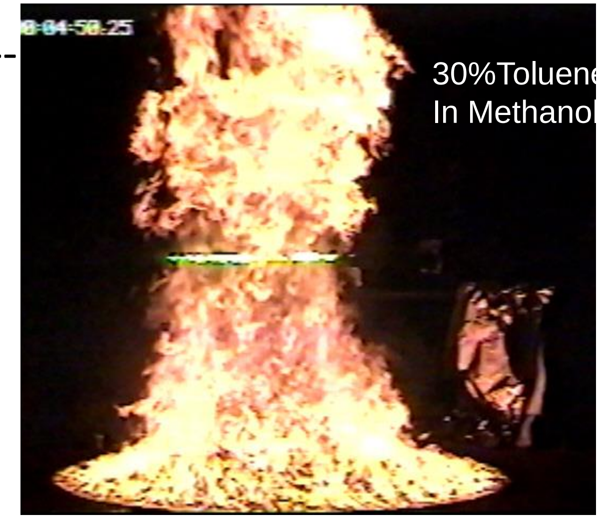
- **Emission term**
  - Temperature
  - absorption coeff → soot *fv*
  - *Local not path integrated*
  - *Key measurement challenge!*

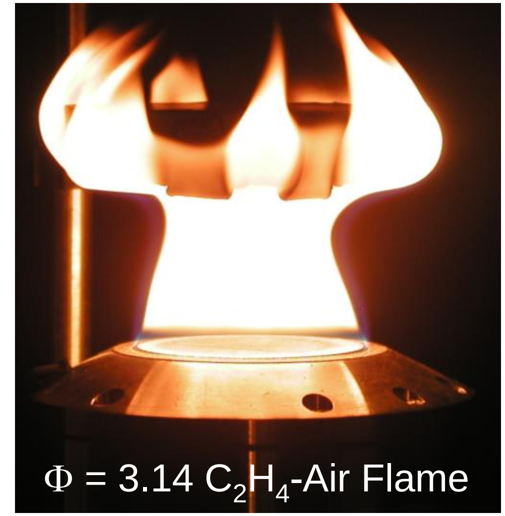
Large-Scale Accident

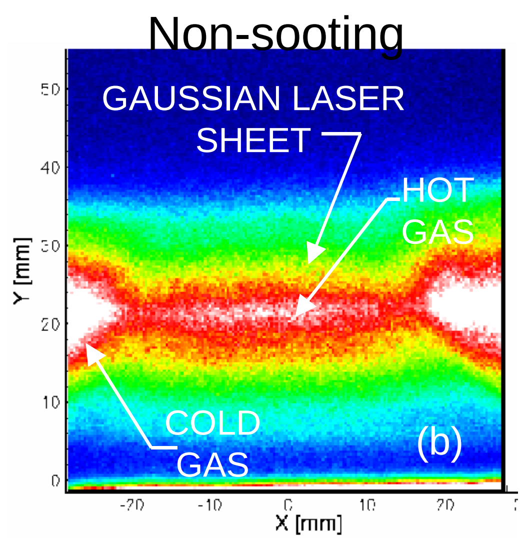
- Technical approach:
  - CARS thermometry
  - LII soot-volume-fraction
  - Fully turbulent fire plume requires meter scales!

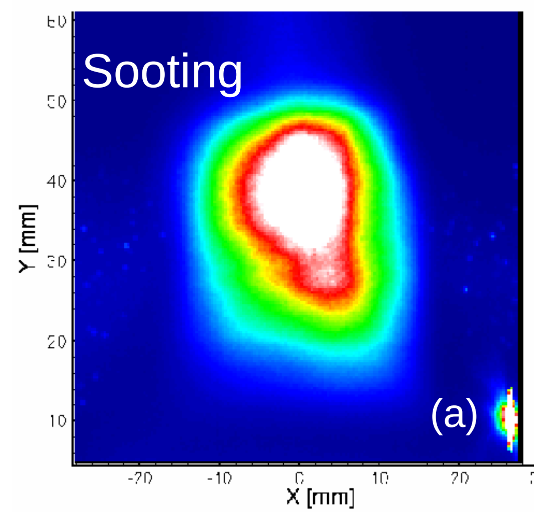
Challenges for Laser Measurements in Fire-Science Experiments

- **Environment:** Experiments are typically performed outdoors --- FLAME Facility
- **Large-scale** of the combustion system
- **Sooting** combustion systems
  - Mie scattering, incadescence, PAH, C_2…
  - Limits choice of optical techniques
- Optical thickness
  - Fiber coupled diagnostics
  - Light pipes
  - Lightly Sooting or Nonsooting Fuels
  - Measurements become more invasive





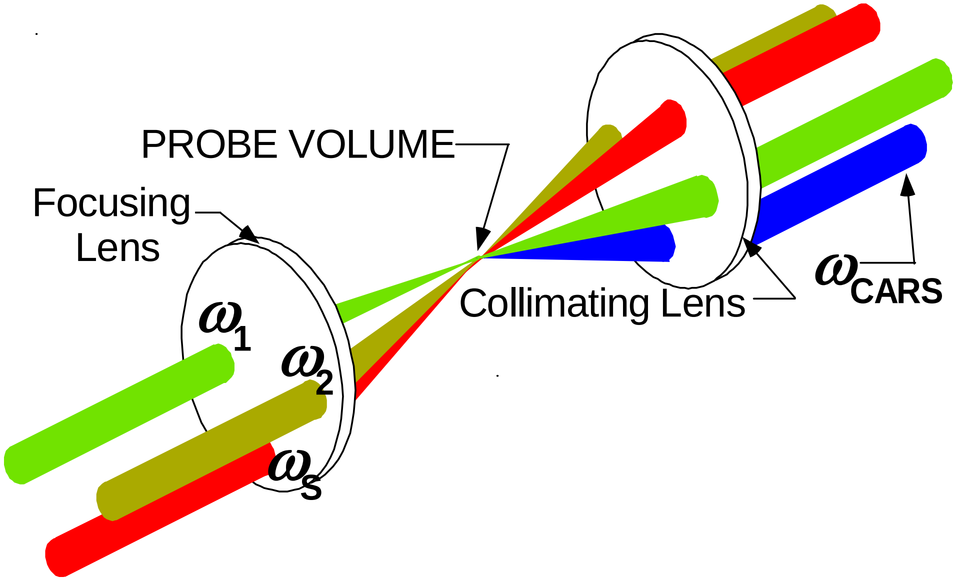


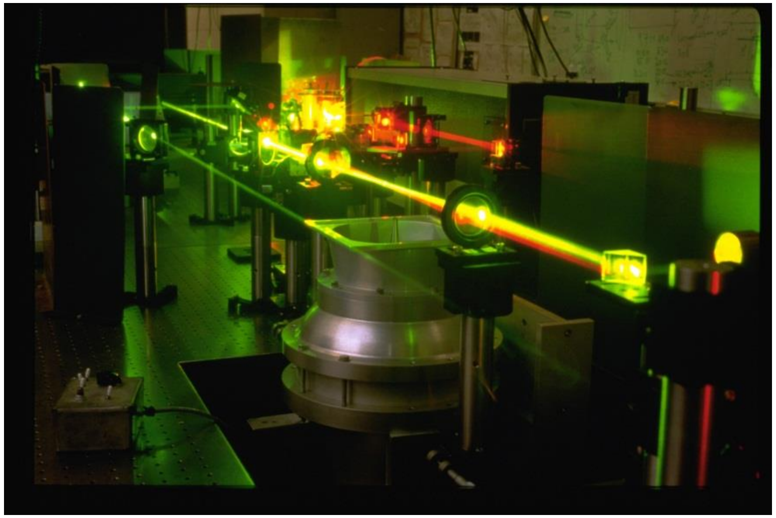


Light-Scattering Images

Kearney *et al.*, AIAA2004-1358.

Coherent anti-Stokes Raman scattering (CARS)

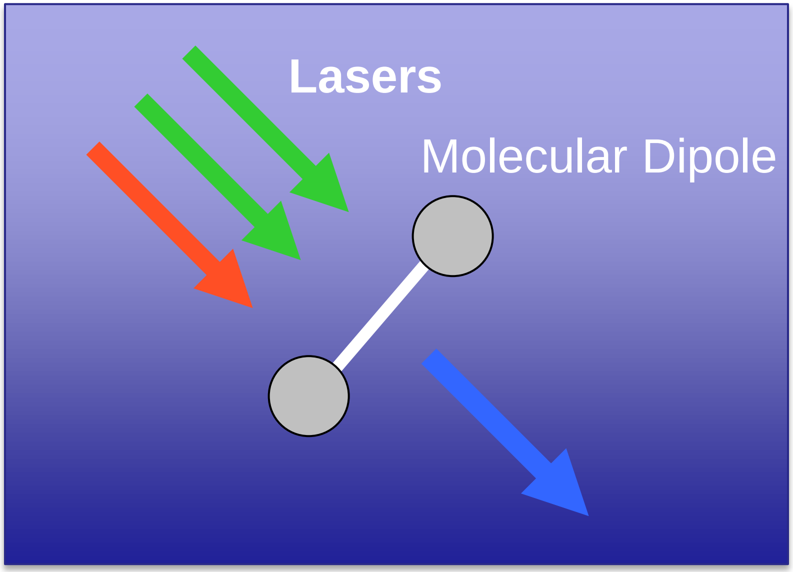




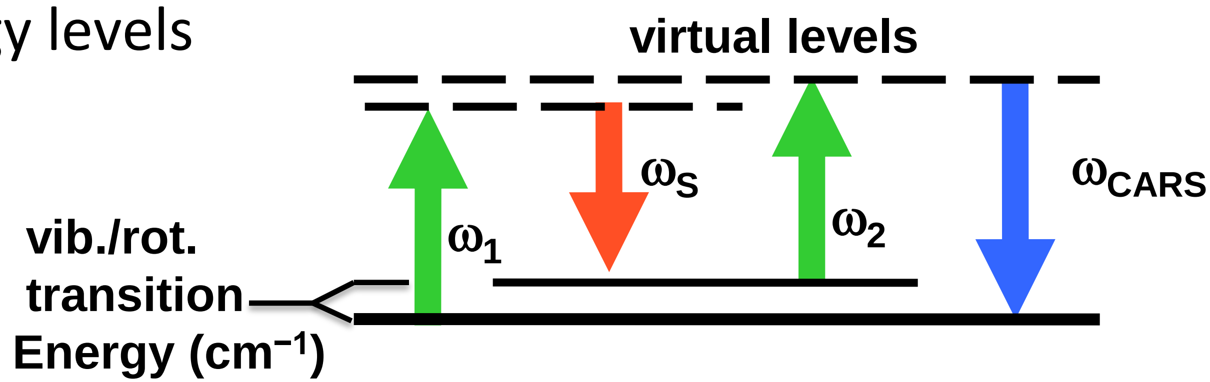
- Three pulsed laser beams (pump #1, pump #2, Stokes) crossed at a common focus
- Signal is generated as a coherent beam within the crossing region
- Signal beam contains information about the vibrational/rotational Raman spectrum of the probed gases
- The signal is blue, or anti-Stokes shifted, away from background luminosity
- Coherent Anti-Stokes Raman Scattering (CARS)

CARS is a coupling of laser light with matter via molecular energy levels

- Three photons interact with the molecule
- The molecule scatters a photon
  - Frequency combination of laser light is tuned to the difference between energy levels



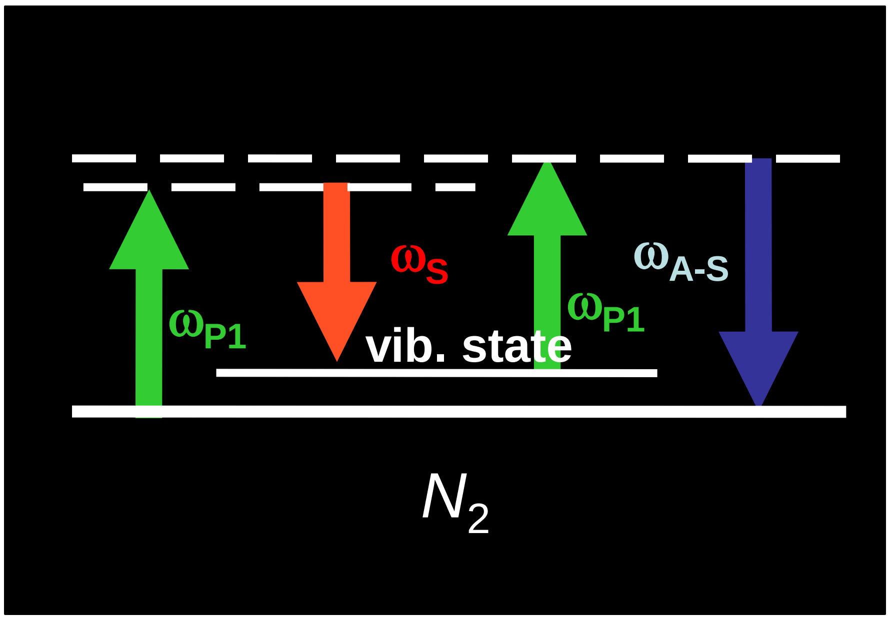
Coherent Anti-Stokes Raman

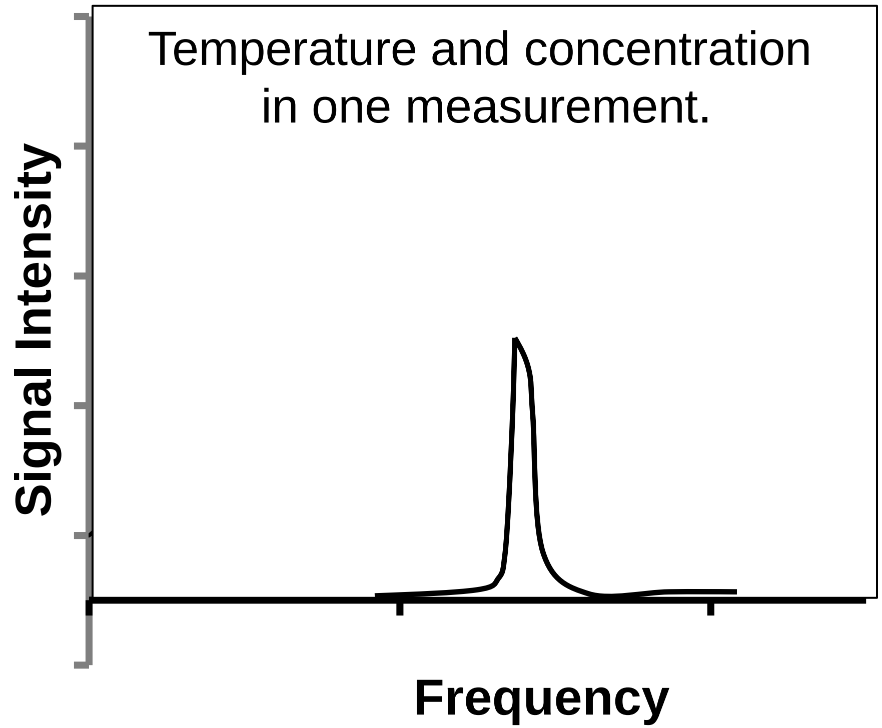


Broadband CARS

Narrowband CARS

- 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

We can work with vibrational Raman or rotational Raman

- CARS spectra reflect temperature via their shape
- Signal for transition from level i to level j

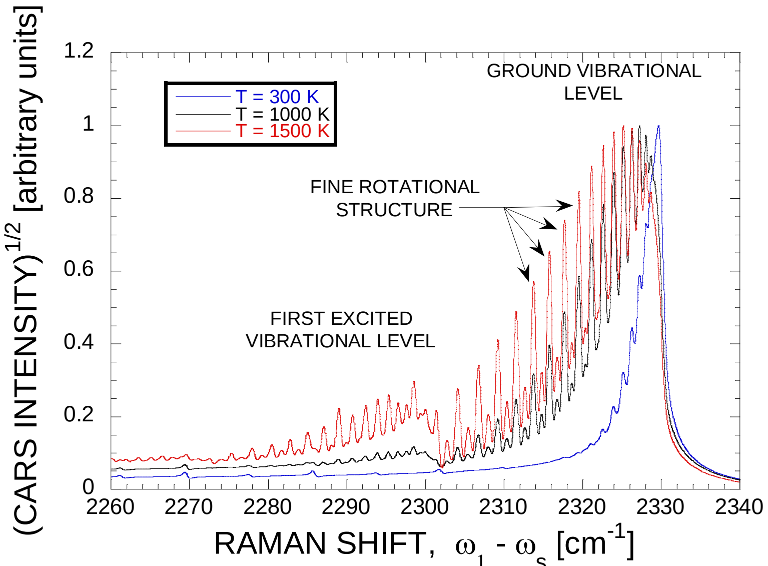
$$I_{CARS} \sim \left(\frac{N_i - N_j}{\omega_R - (\omega_1 - \omega_s) + i\Gamma} \right)^2$$

  - Spectral shapes reflect internal energy partitioning via Boltzmann distribution

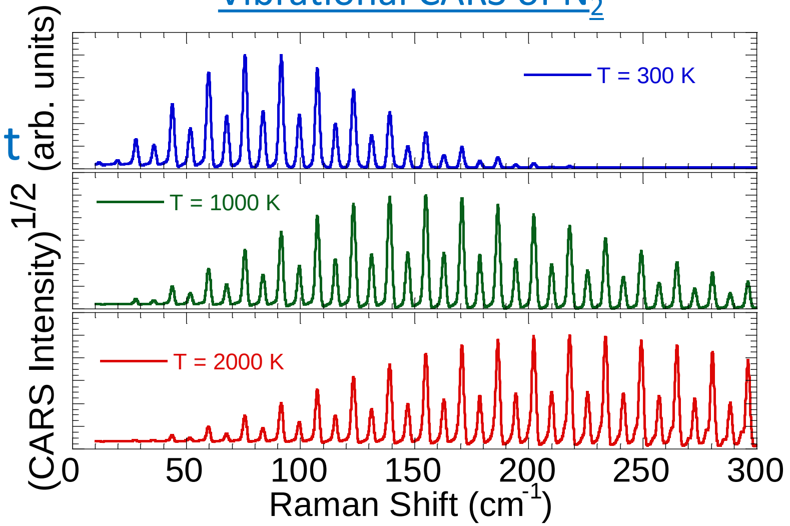
$$\frac{N_i}{N} = \frac{g_i \exp(-\varepsilon_i / kT)}{\sum_i g_i \exp(-\varepsilon_i / kT)}$$

  - We measure the CARS spectrum and then fit the data to theoretical prediction

- Measured parameters include:
  - Temperature: 2% accuracy 1-4% precision
  - Species
  - Pressure



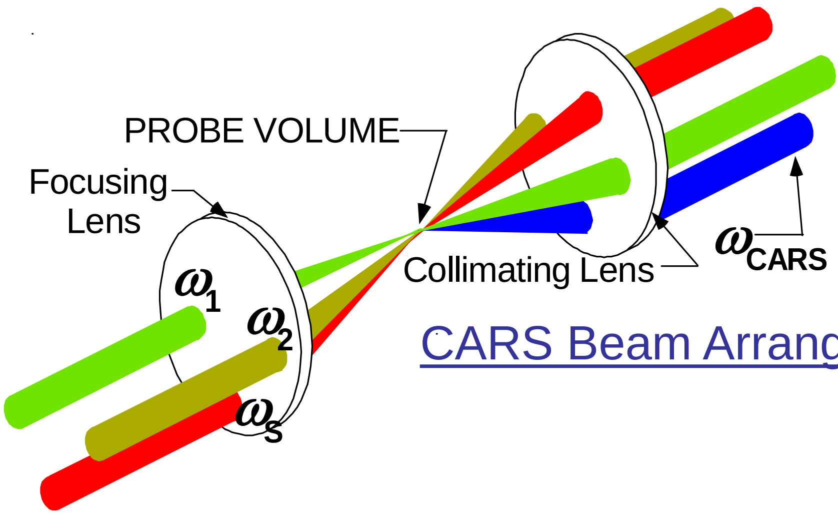
Vibrational CARS of N_2



Rotational CARS of N_2

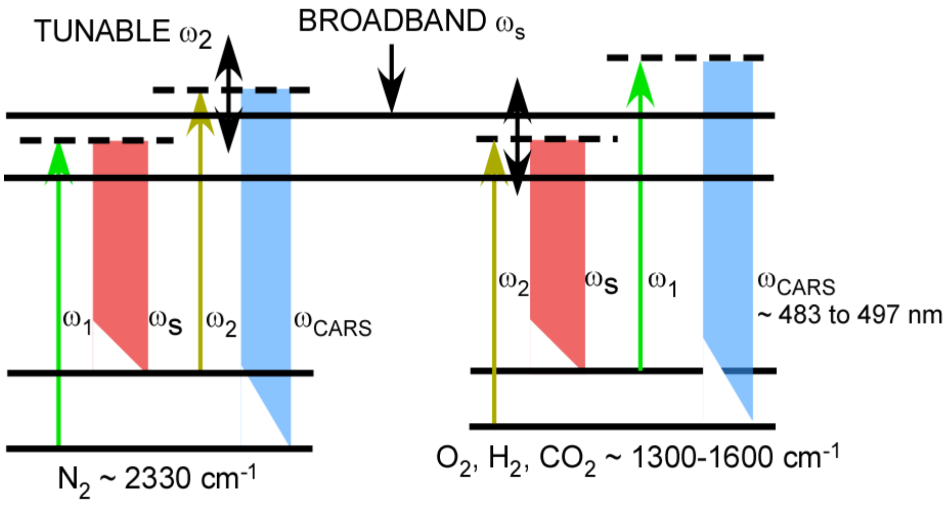
Dual-Pump CARS Approach for Spatially Resolved Pool-Fire Thermometry

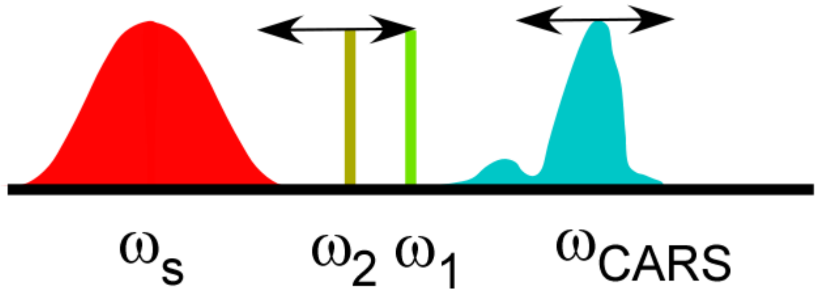
- Coherent, laser-like signal
  - Spatial discrimination, slow collection optics
  - **<u>Readily coupled to optical fibers</u>**
- Blue-shifted signal permits spectral discrimination against red/IR flame emission
- Spectrally tunable output positions CARS signal beam in “quiet” regions of the spectrum away from C_2 interference



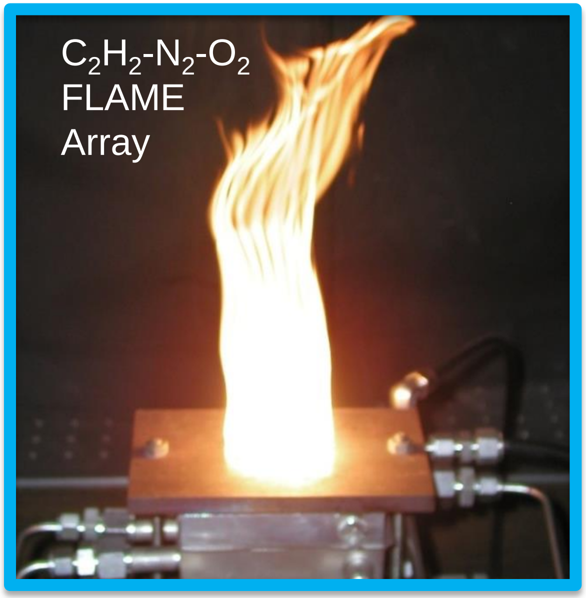
CARS Beam Arrangement

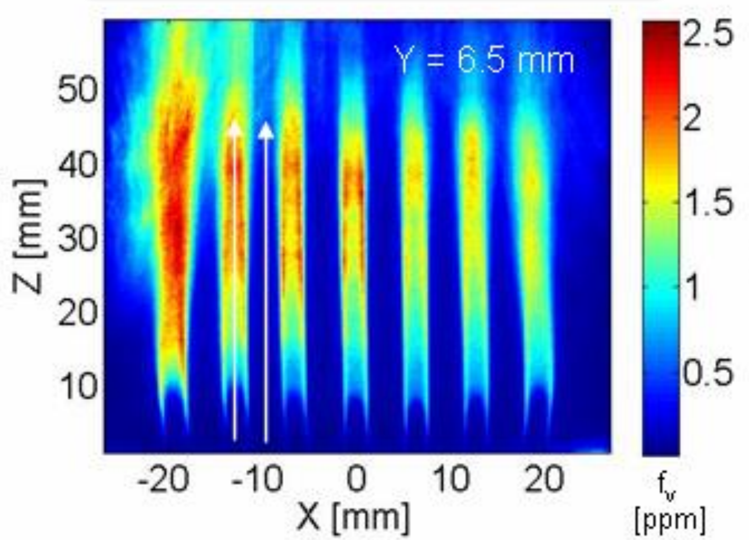
$$\omega_{CARS} = (\omega_1 - \omega_s) + \omega_2$$





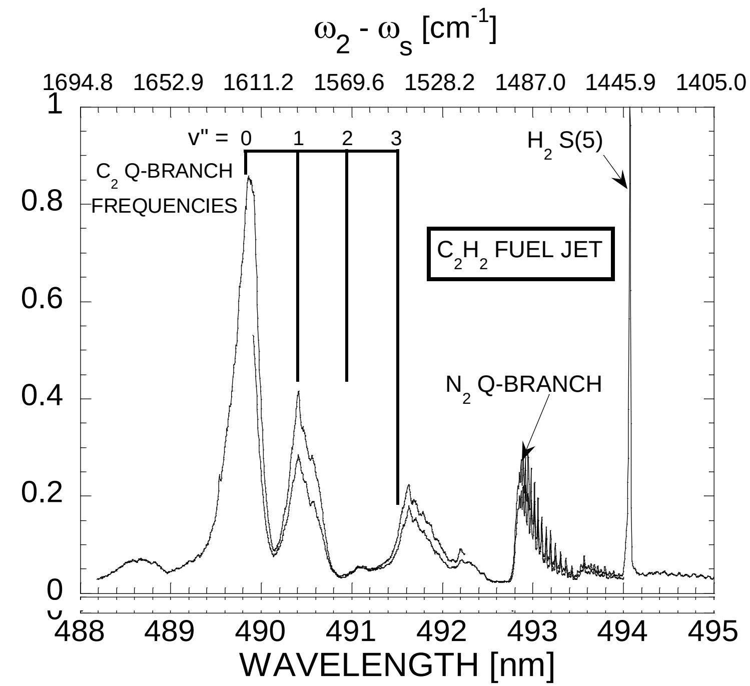
Laboratory Measurements in C_2H_2-fueled Diffusion Flame Array





Soot Volume Fraction by LII

- Initial DP-CARS measurements performed in laboratory scale burners
- "How much soot can DP-CARS handle?"
- Tune pump-2 frequency to find "quiet" spectral regions
- Observe levels of C_2 Swan band interference in flames with ppm-level soot



Results from Heavily Sooting C_2H_2 Fuel Jet

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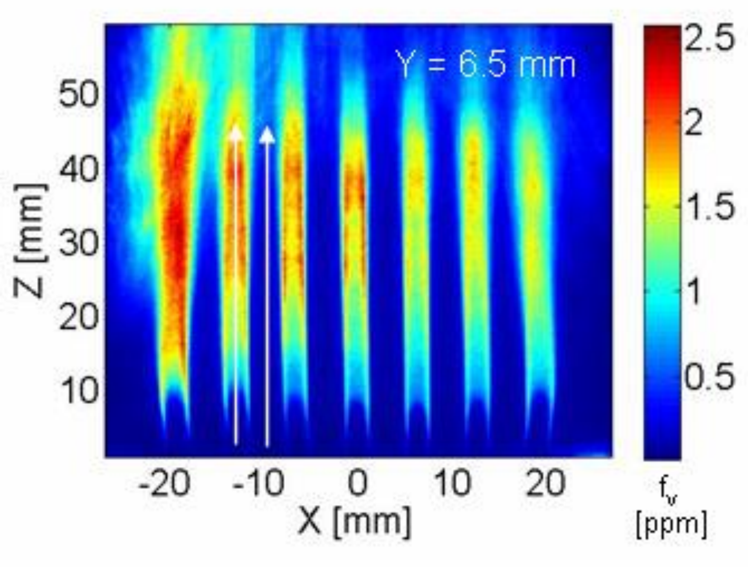
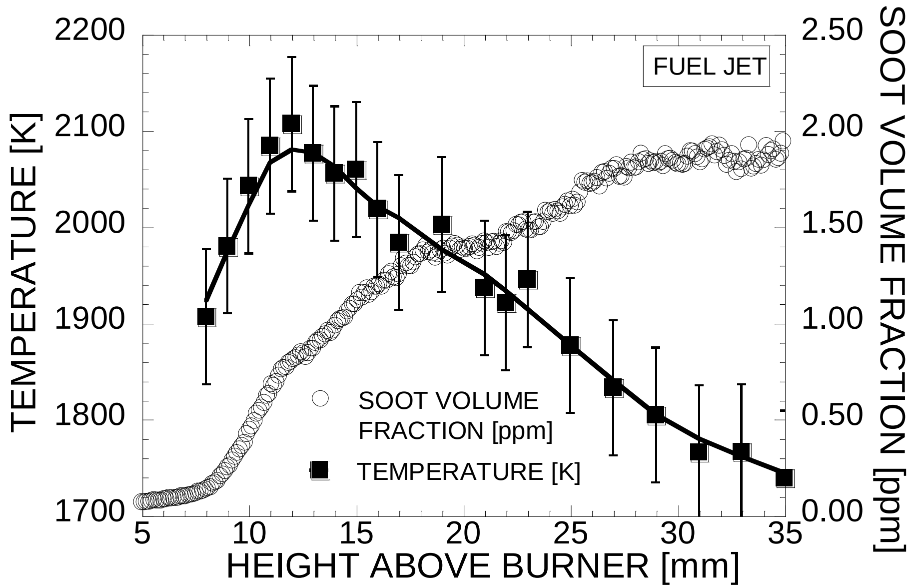
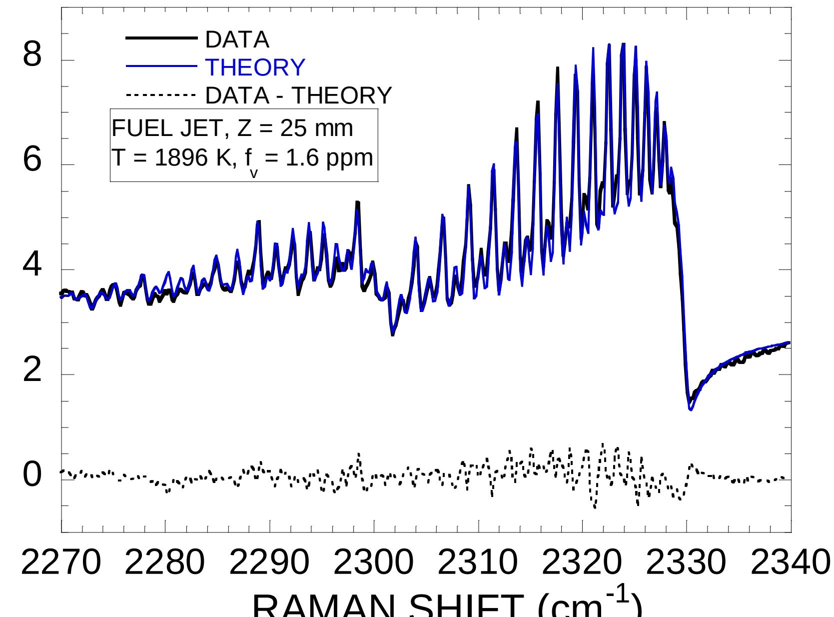




- Fits performed using Sandia CARSFT code
- Both N_2 and H_2 Included in the Dual-Pump theoretical calculations due to close proximity of H_2 rotational lines to N_2 Q-branch
- Combustion product susceptibility used in O_2/N_2 stream and above Al. feed port
- Acetylene susceptibility used in Fuel Jet
- Temp and N_2 mole fraction floated in fitting procedure

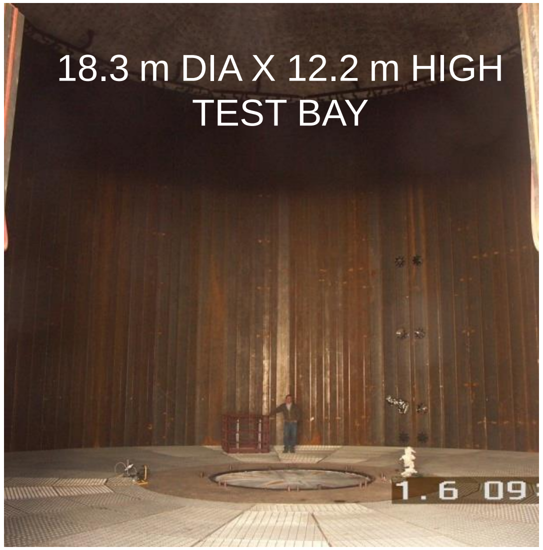


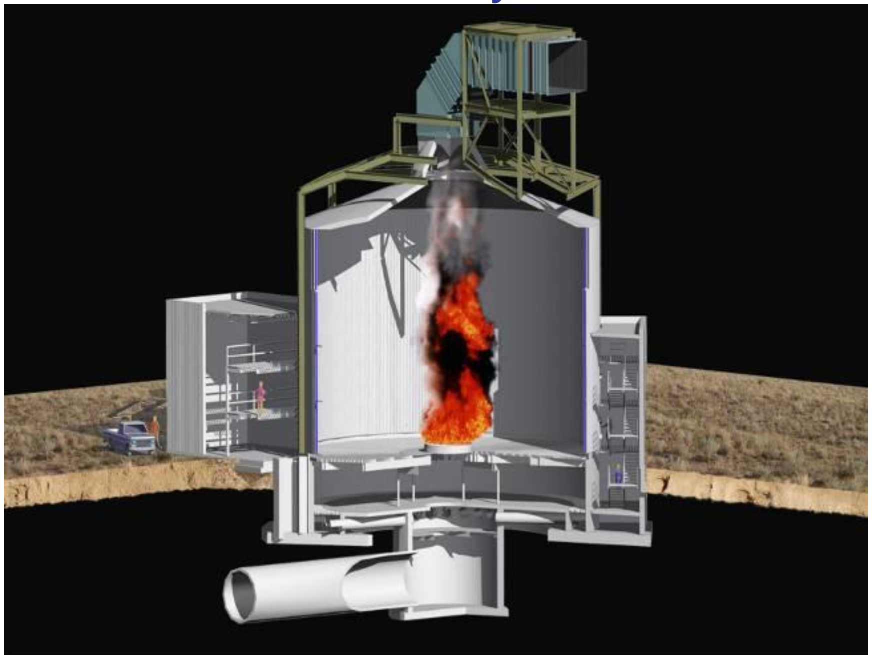
Next Step: apply diagnostic for pool-fire measurements

Kearney and Jackson, *AIAA J.* **45,** 2947, 2005.

FLAME Facility Enables Full-Scale Testing with High-Fidelity Laser Diagnostics

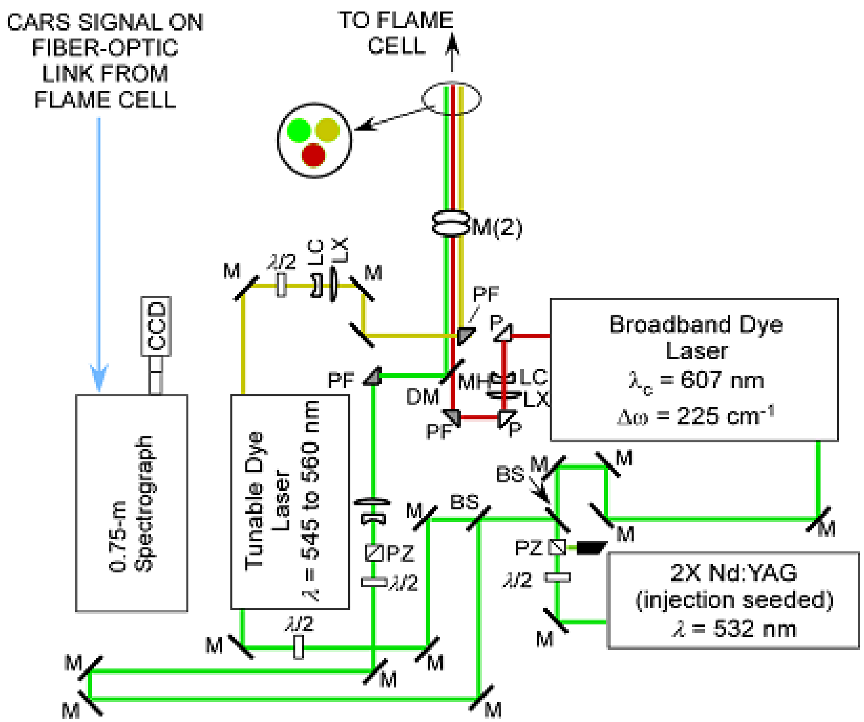
- Fire Laboratory for Accreditation of Models and Experiments (FLAME) Facility
- Designed to facilitate deployment of optical diagnostics for full meter-scale fire testing
- Optical Access Ports and Adjacent Lab Space at East,West, and South Positions Around Test Cell
- Liquid and Gas-Fueled Fires up to 3-m in Base Dia.
- Brings Laboratory Control to Full Scale Testing
- Canonical Wind-Free Buoyant Fire Plume

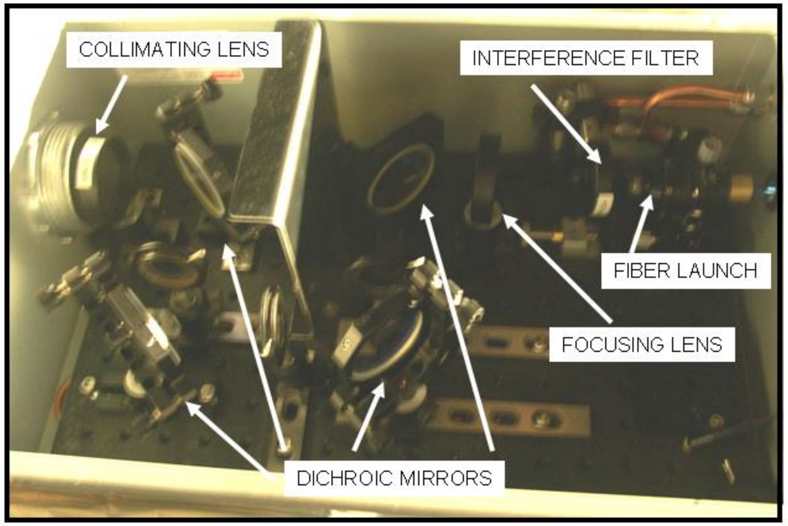




Dual-Pump CARS Instrument at FLAME

- First-ever implementation of CARS for large-scale fire testing
- Methanol and sooting methanol/toluene blends have been tested to date
- Simultaneous mole-fraction measurements have been added to thermometry capabilities





Results – Temperature and Species Data from a Methanol Pool Fire

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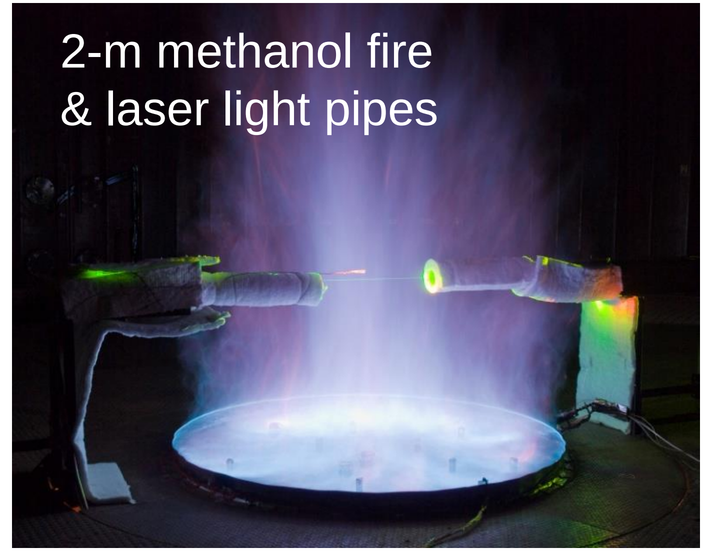
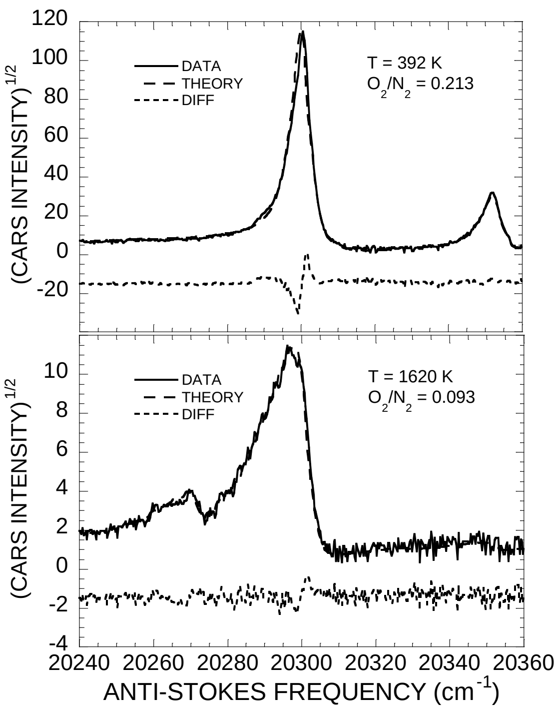
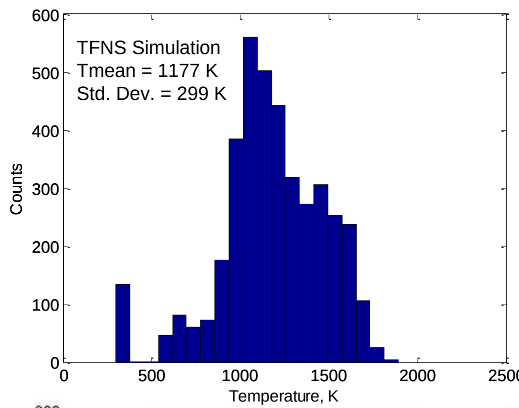
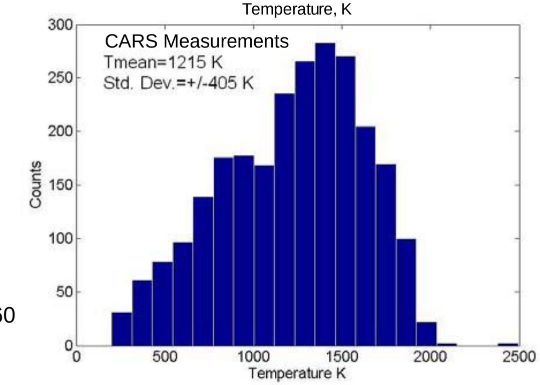
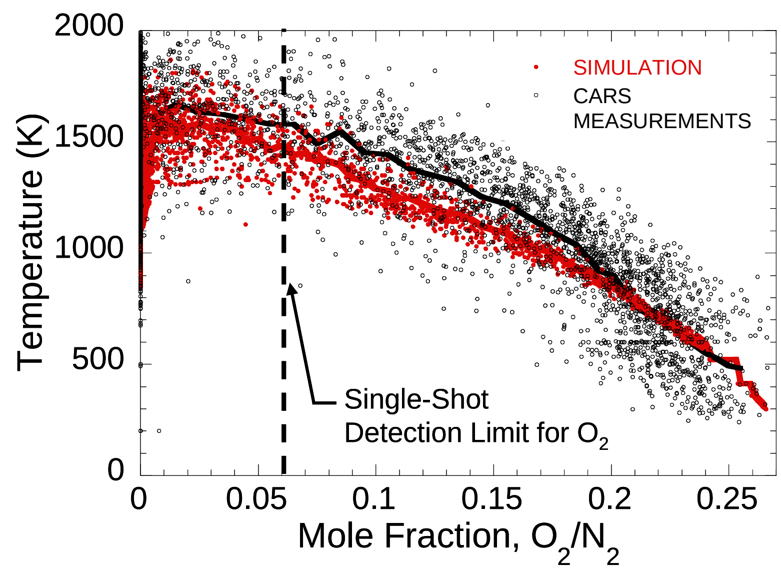
- First experiments conducted in methanol fire
- Nonsooting fuel is simpler starting point for diagnostic development
- N_2,O_2, H_2 observed in DP-CARS spectra
- Temperature and simultaneous O_2 data (ref. to N_2) extracted











Frederickson, Kearney et al., Combust. Sci. Technol., 2009

Laser-Induced Incandescence (LII)

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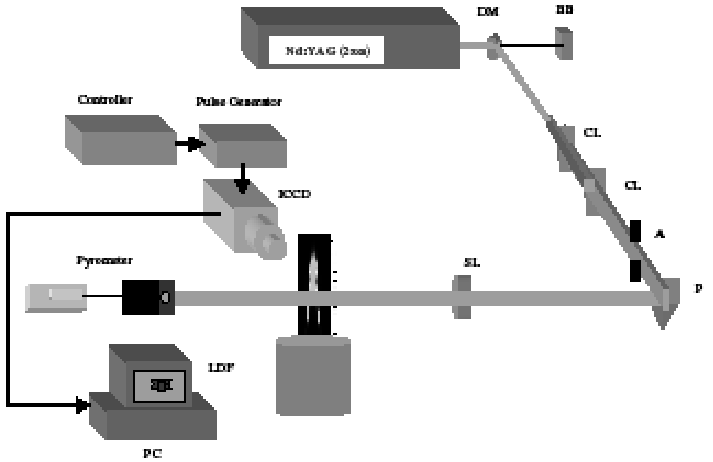
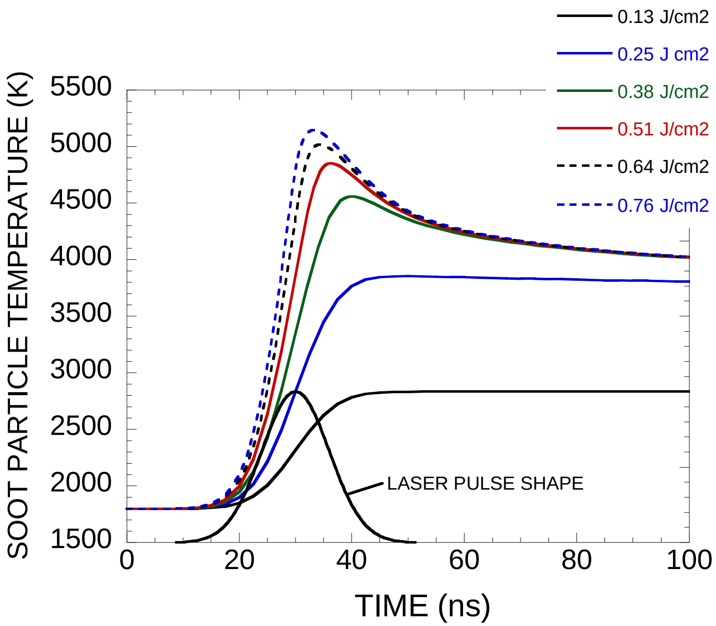
- *in situ* measurement of soot volume fraction
- Soot particle size also possible with ns time resolved LII (TIRE-LII)
- Soot particles heated to vaporization point (4000-5000 K) by absorption of laser radiation
- Thermal emission from soot is monitored during and/or after laser pulse
    - Laser-heated soot emits in excess of background
    - Total emission ~ soot volume
    - Time decay of emission ~ d_p
- 2-D imaging
    - ns time resolution
    - 50-100 μm spatial resolution
- Calibration against light extinction or other known particle dist. is required



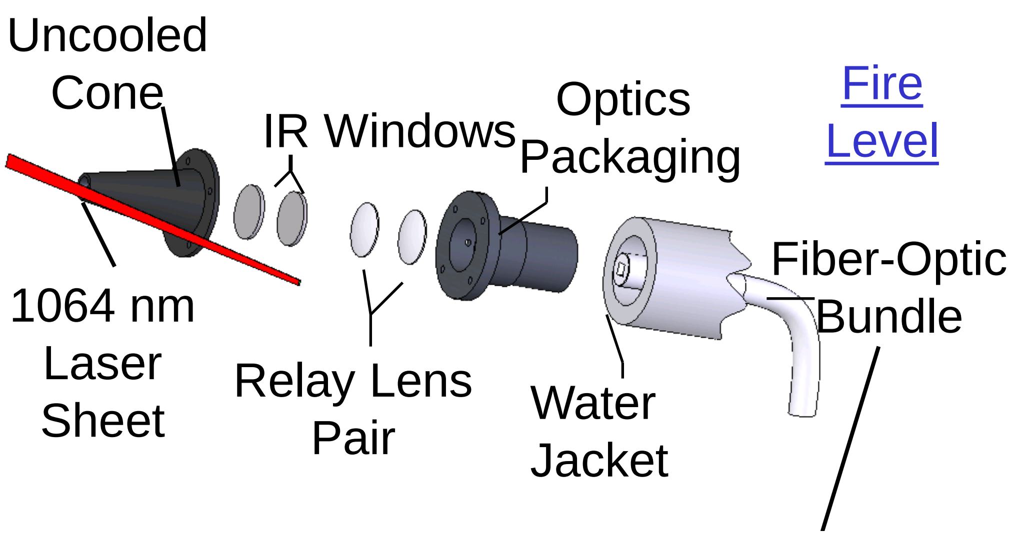
LII Experiment (Santoro and Shaddix, 2002)



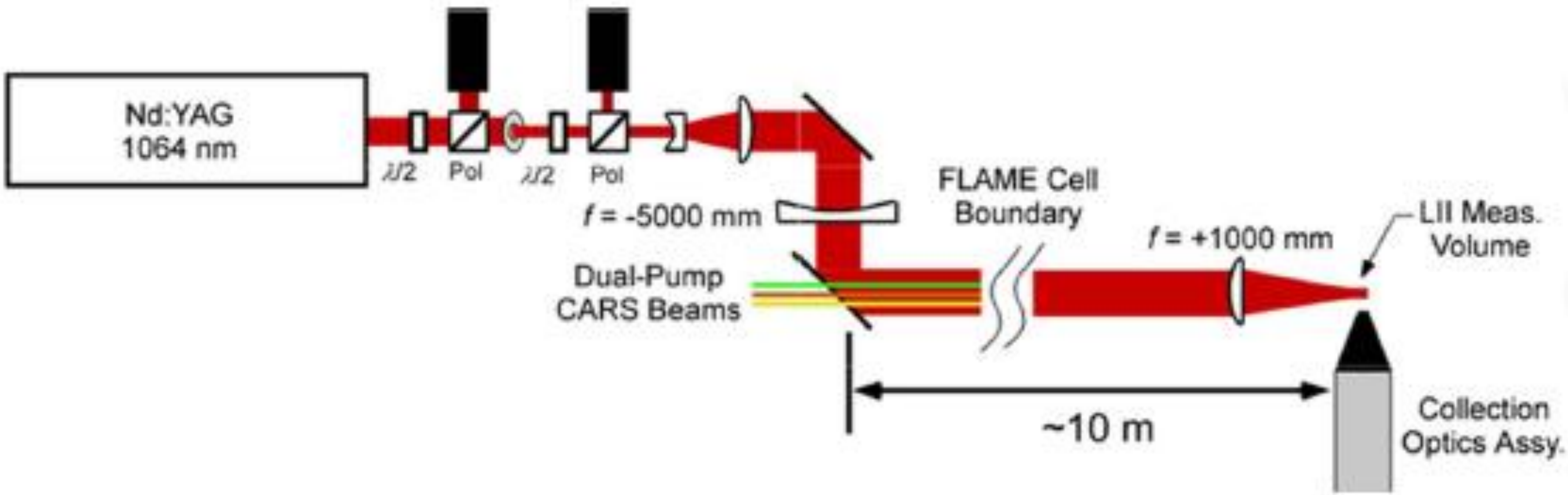
Soot Particle Temperature Rise

Laser-Induced Incandescence Instrument at FLAME

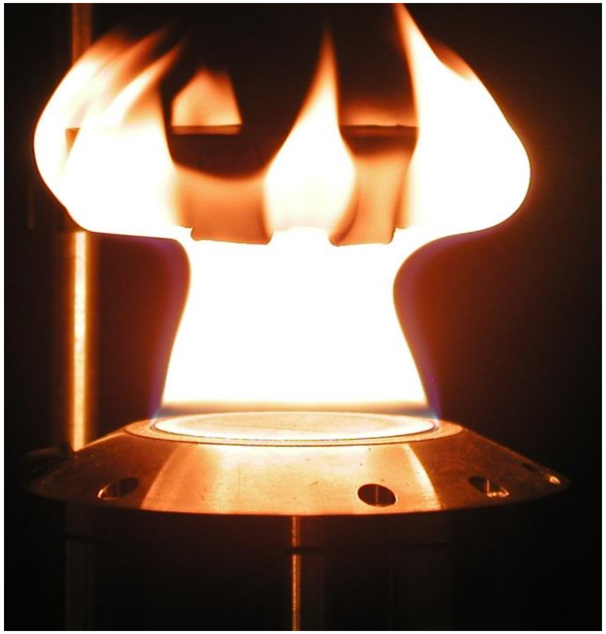
- Toluene/Methanol blended fuel, ranging from 10/90 to 30/70 ratio, higher sooting propensity than pure methanol
- 1064nm laser sheet, fluence ~ 1.14 J/cm^2
- LII probe, water-cooled jacket, 200mm achromatic relay lens pair, 1cm^2 fiber-bundle (10 μm) couples signal to ICCD camera
- Four stage automated translation system to position probe and adjust focus
- Signal is filtered using IR-reflecting window gating reduces background emission from fire

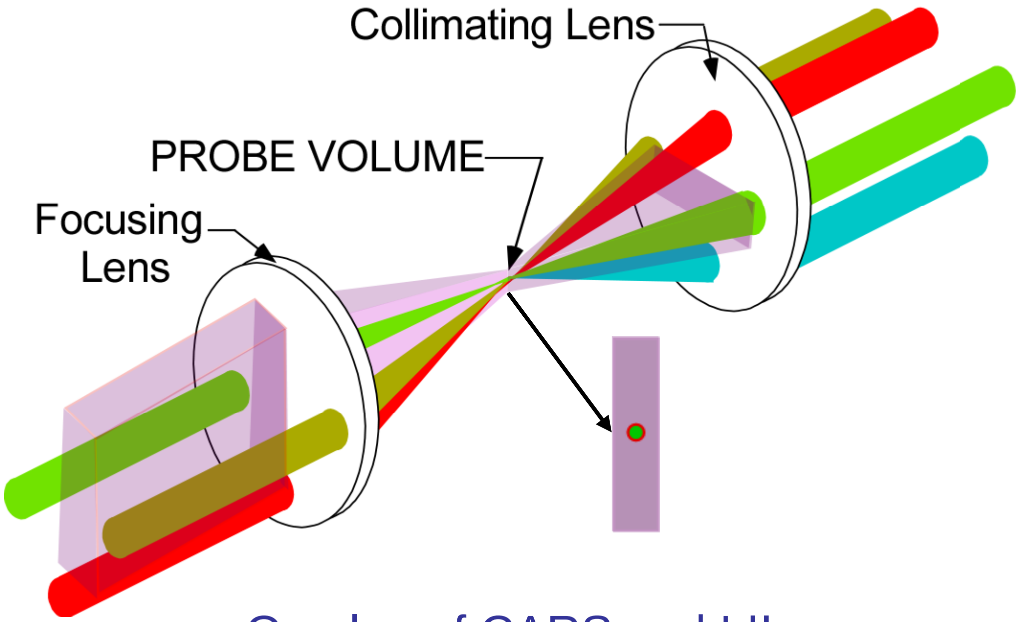


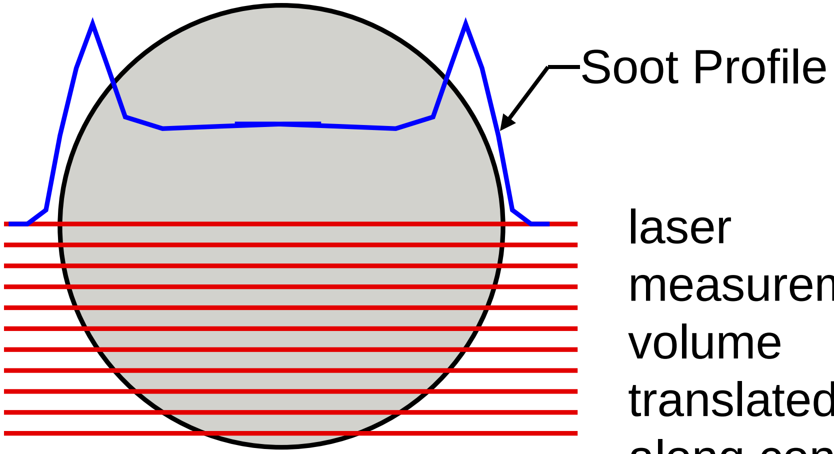
[Fire Level]



Joint CARS/LII Measurements Performed in Premixed C2H4/air Flame

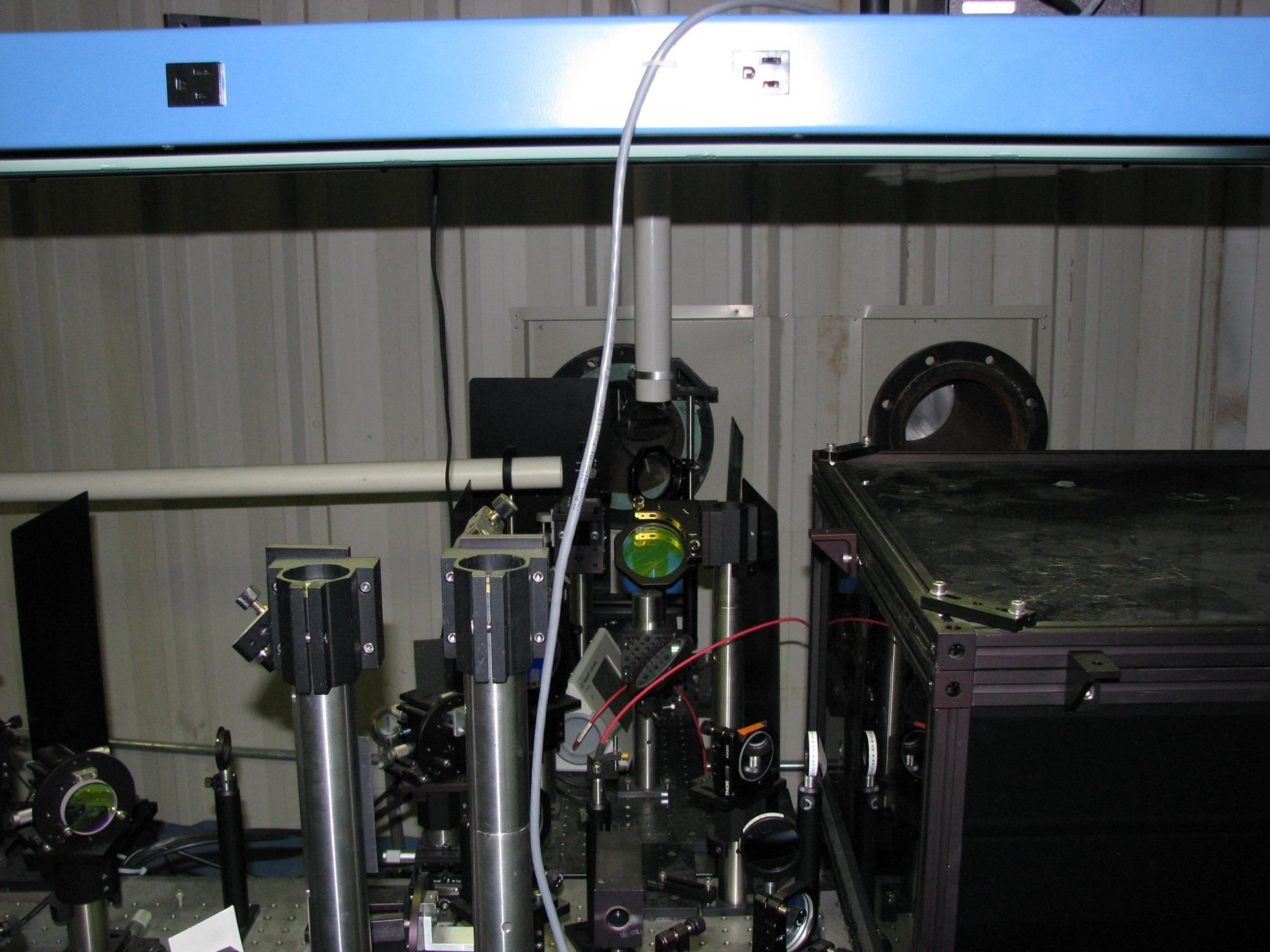






laser measurement volume translated along constant-height planes

- Overlap of CARS and LII beams verified using laser burn paper
- LII detection system pointed at CARS probe volume by imaging pump-beam overlap
- CARS/LII volumes translated through gradient region and across burner and constant height



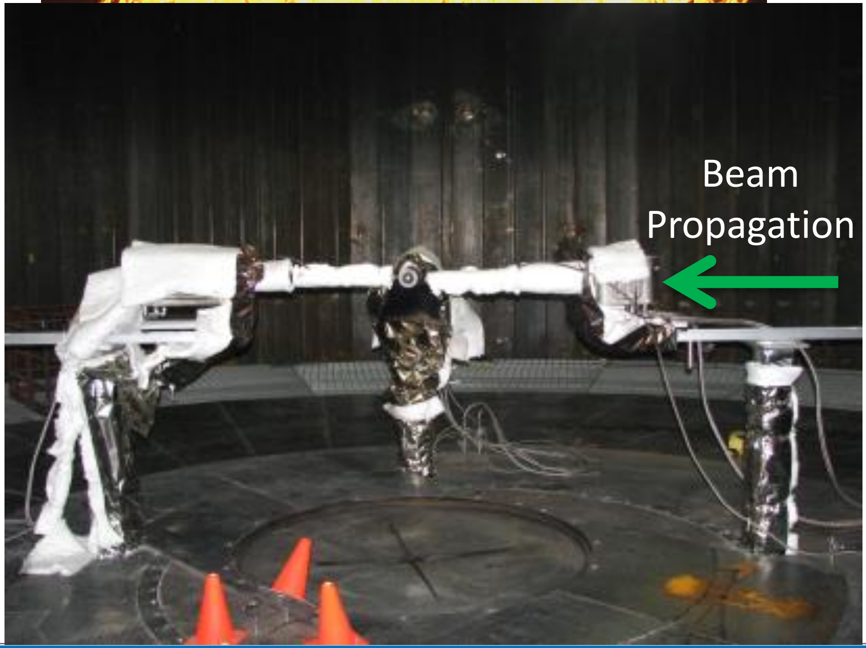
Profile Measurements in 10% Toluene Research Fuel Blend

- 10% toluene added to methanol
- Data recorded along radial profiles at 3 heights: 0.5, 1.0 and 1.5 m
- Encompasses “vapor dome” and actively burning regions
- Constant fuel level for 1-2 hr burns
- Measurements begin at plume center with highest possible CARS signal
- Proceed radially outward – compensates for signal loss due to fiber thermal misalignment
- 3000-5000 single-shot CARS/LII realizations before moving measurement volume
- Several different burns at each height for ensemble averaging

y = 1.5 m

y = 1.0 m

y = 0.5 m



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

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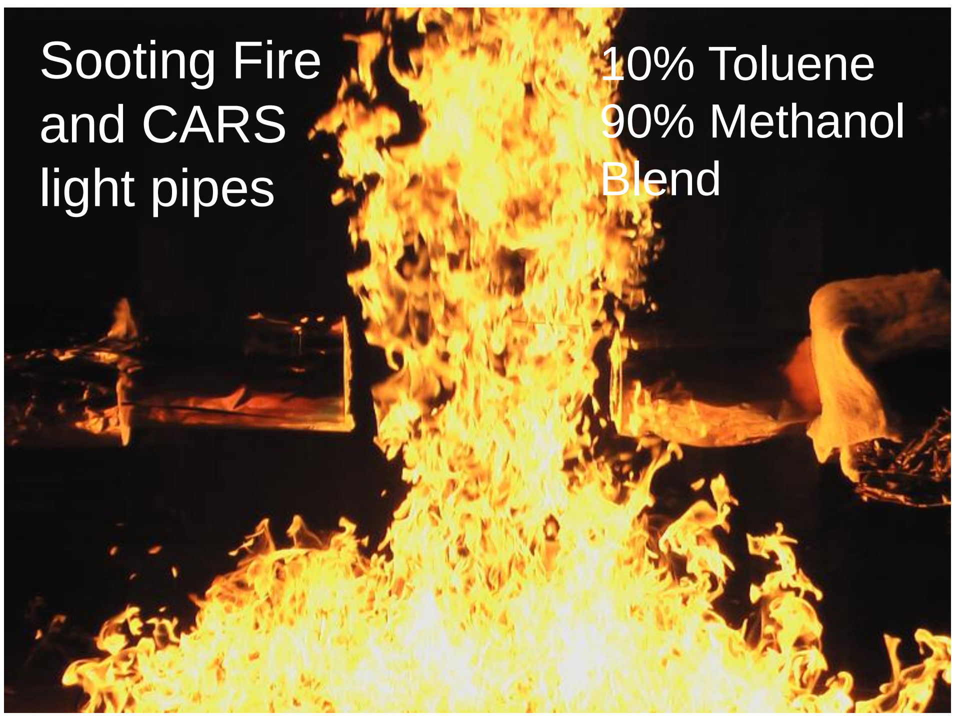
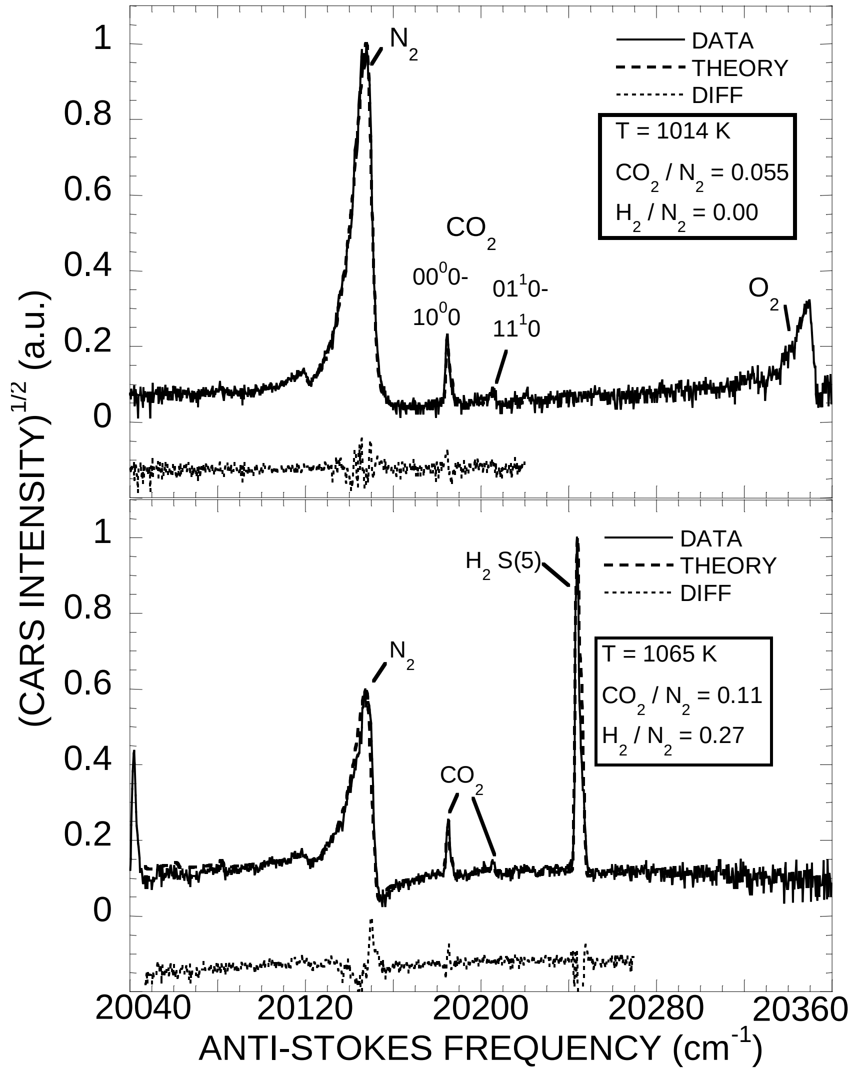
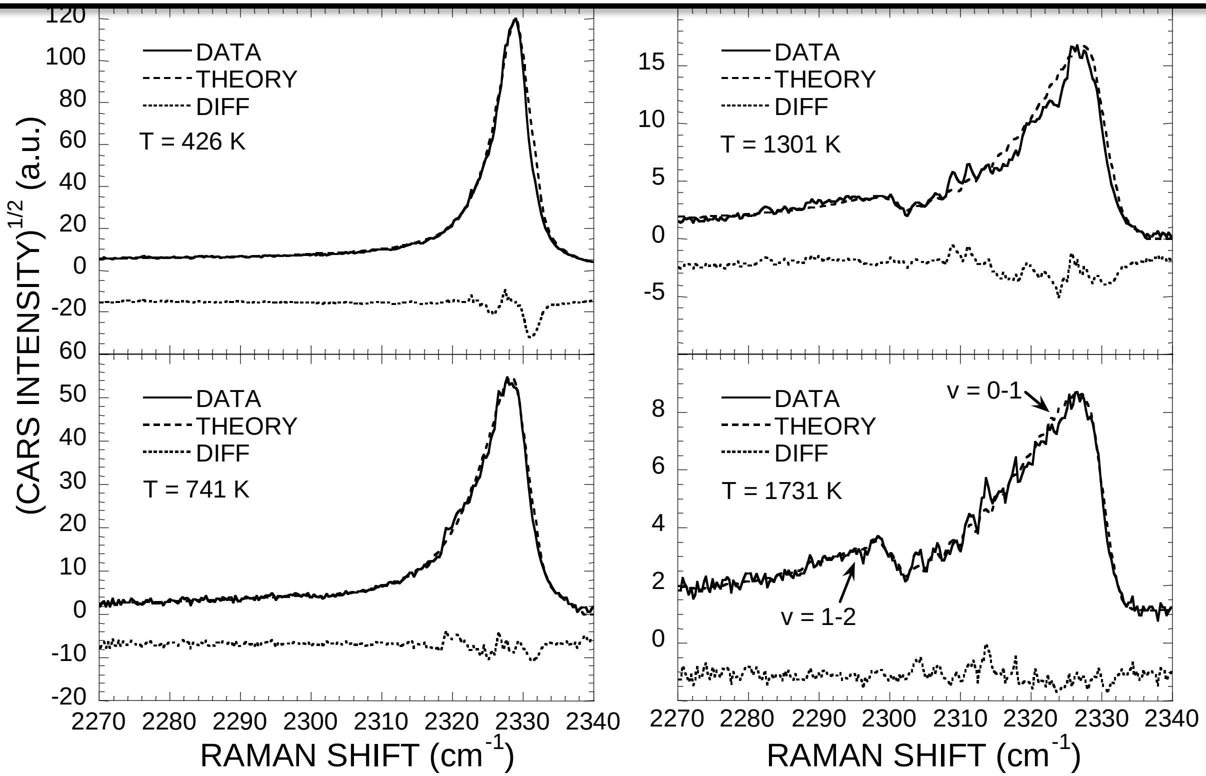
- CARS spectra from sooting fire show N_2, CO_2, H_2, and O_2
- Full ensemble of species data not available as of yet
- Two representative spectra with theoretical fits (Sandia CARSFT code) shown here

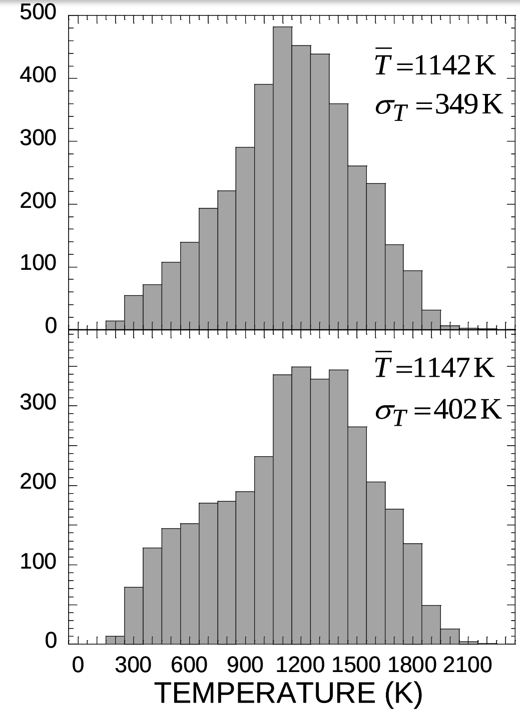




Temperature Measurements in Sooting Pool Fire



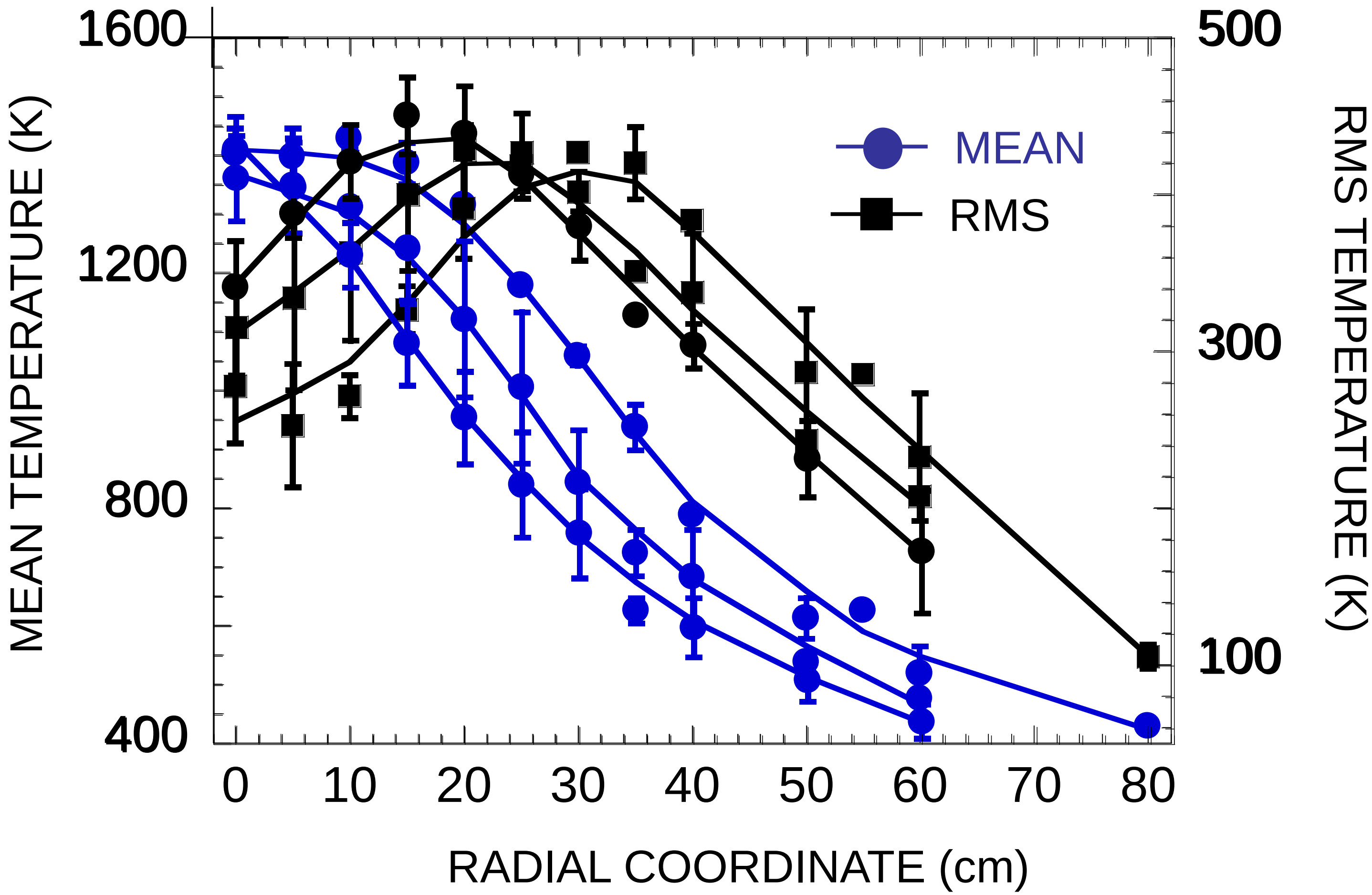
Single-Laser-Shot Spectra and Theoretical Fits



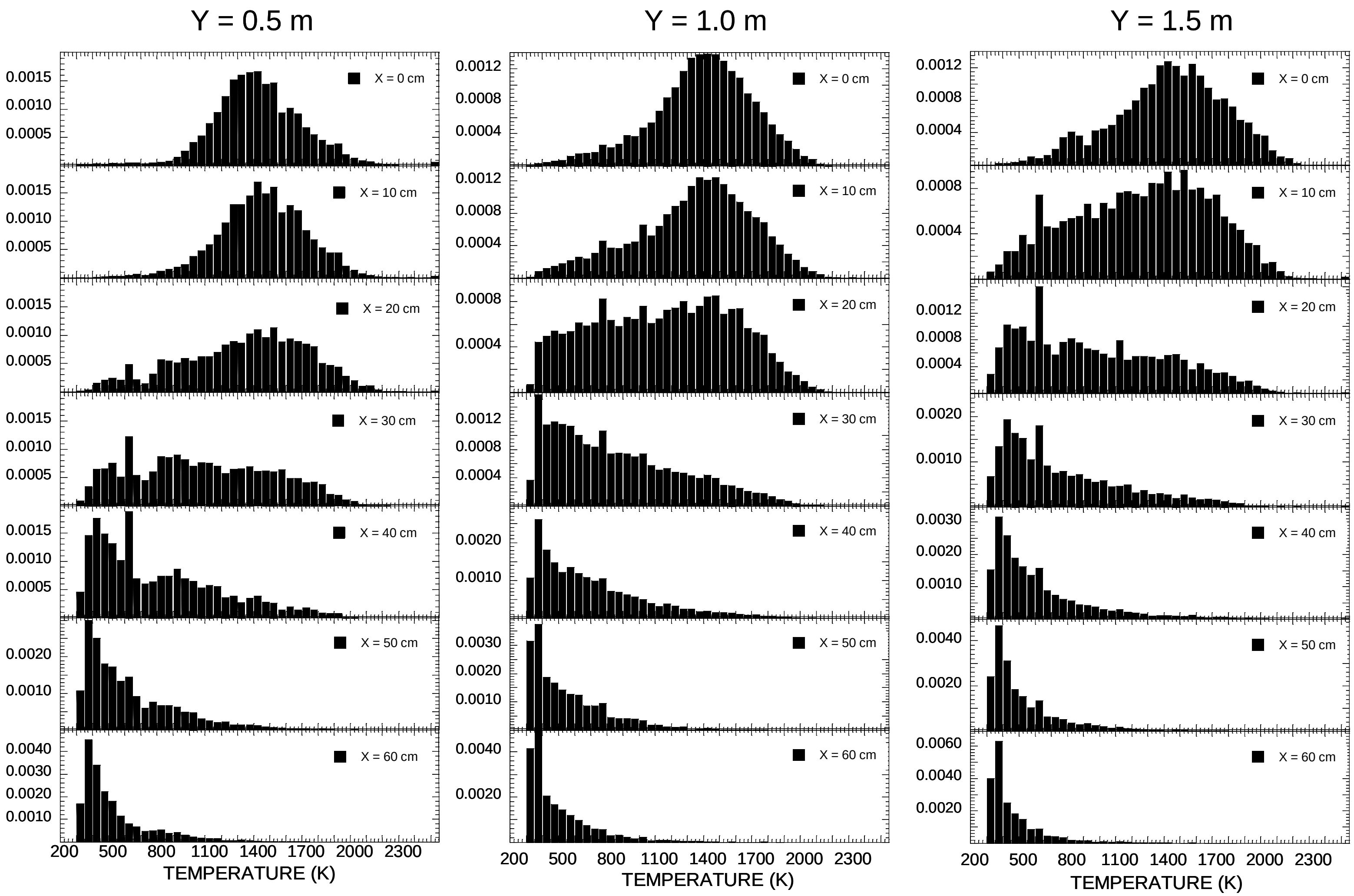
Temperature PDFs from 2 Separate Methanol/Toluene Burns

- CARS is applied in a pool fire with a representative volume loading of O(0.1 ppm)
- Measurement volume is 0.5 diameters above the center of the 2-m fuel pan
- 3500 single-shot temperature realizations per burn used to construct pdfs
- Wide std. dev. indicative of large-scale turbulent mixing in pool fire
- Mean temperatures compare favorably with nearby TC reading 1210 K

Radial Temperature Profiles

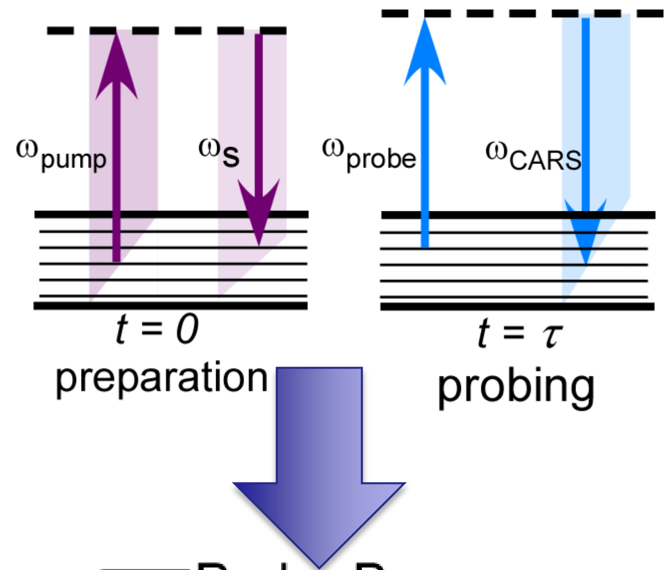


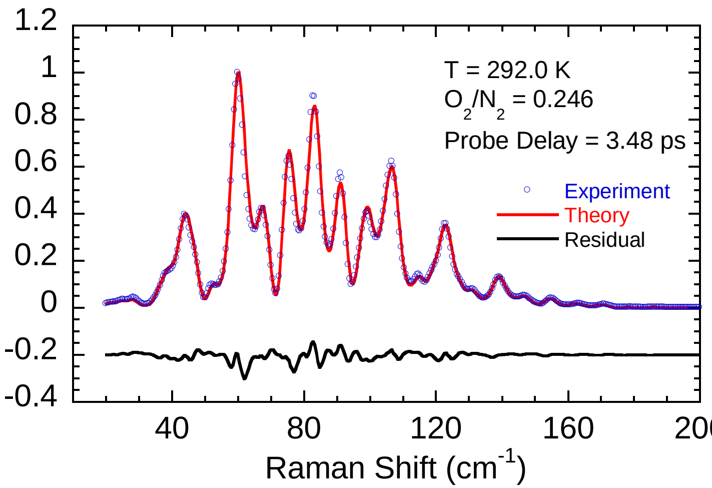
Temperature PDF

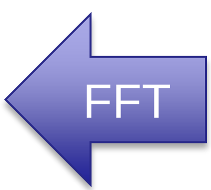


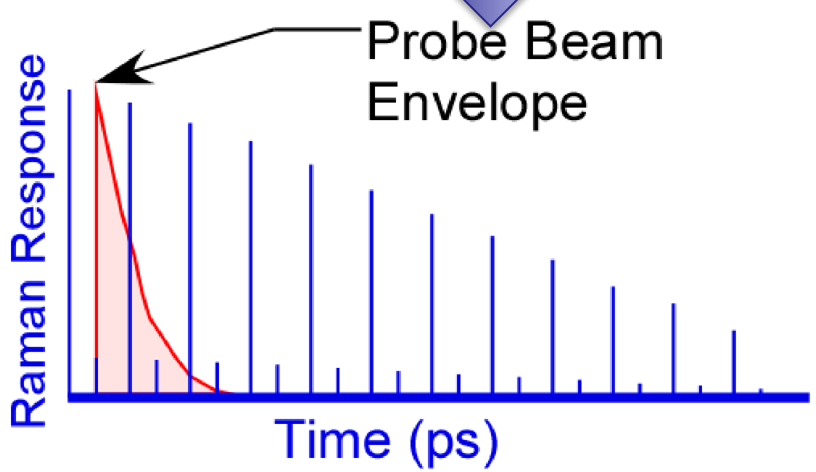
Femtosecond CARS

- Why develop fs CARS when ns CARS works pretty well?
  - Precision: 3-4× improvement in thermometry
  - Data rate: kHz data rates as opposed to 10 Hz
  - Time resolution: 10 ps or better (shock physics)
  - High-pressure environments
  - Background-free measurements
  - Particle-laden environments





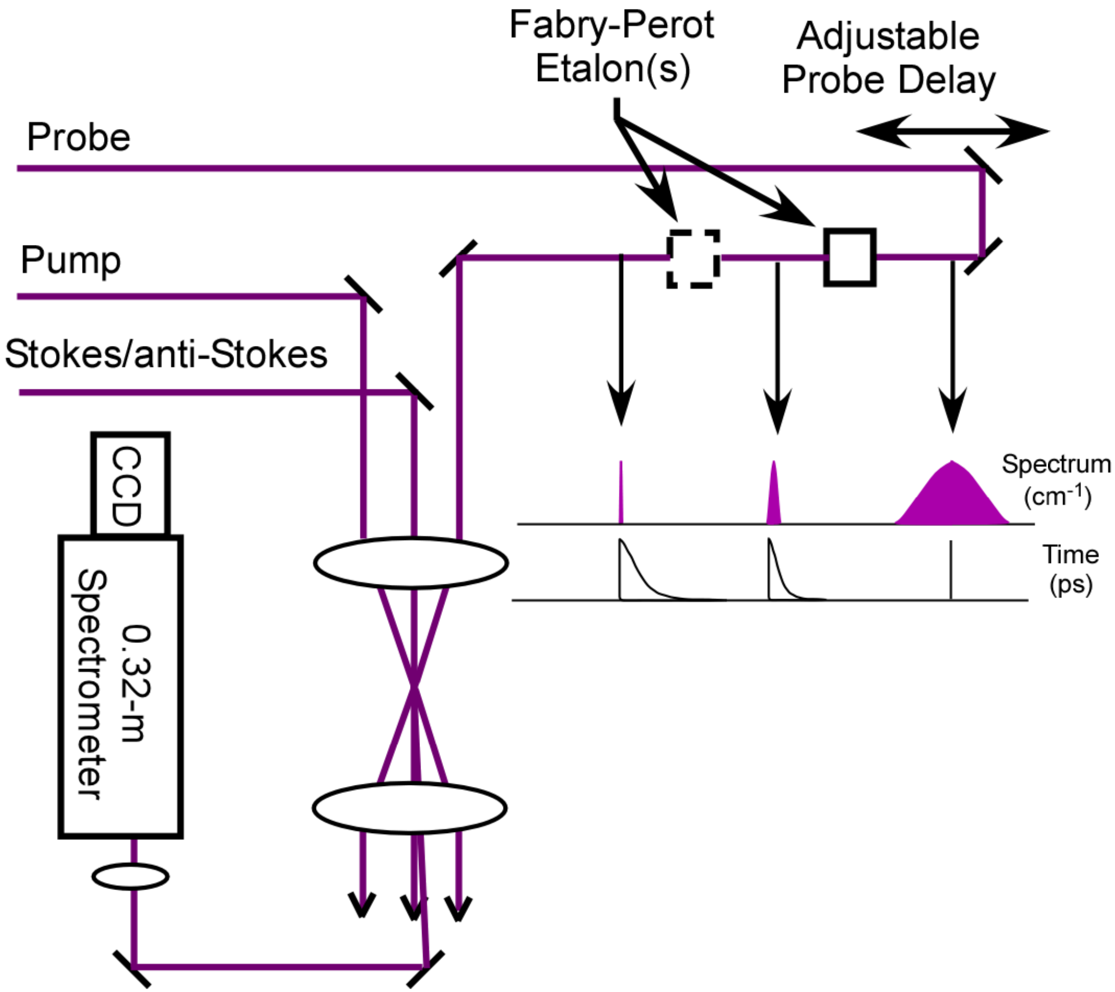




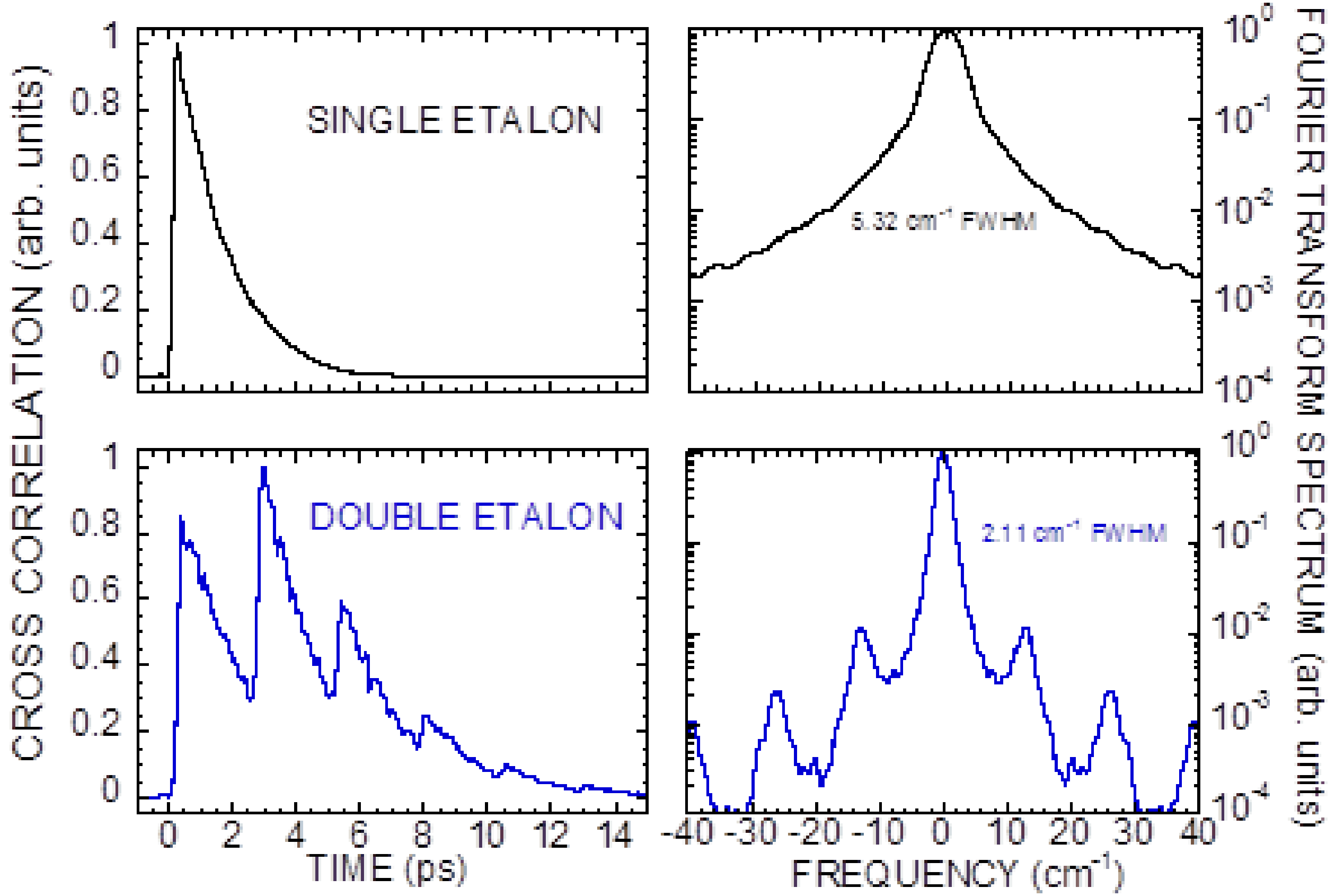
Detect in the frequency/spectral domain

Model in the time domain

Sandia femtosecond CARS system



Probe beam E-field envelope measurements



A simple phenomenological model is implemented in Matlab for spectral fitting

- The time evolution of the molecular Raman response is calculated from

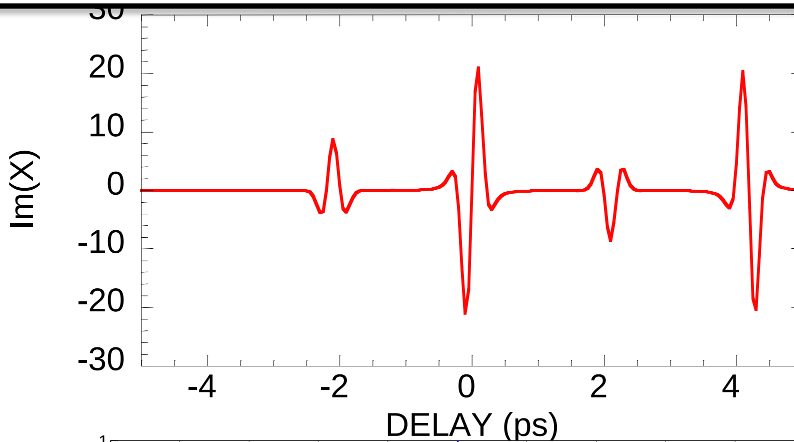
$$\chi(t)=\sum_{k}\sum_{\Delta J=\pm 2} W_{JJ'}^{(k)}\exp\left(i\,\omega_{JJ'}^{(k)}t\right)\exp\left(-\Gamma_{JJ'}^{(k)}t\right)$$

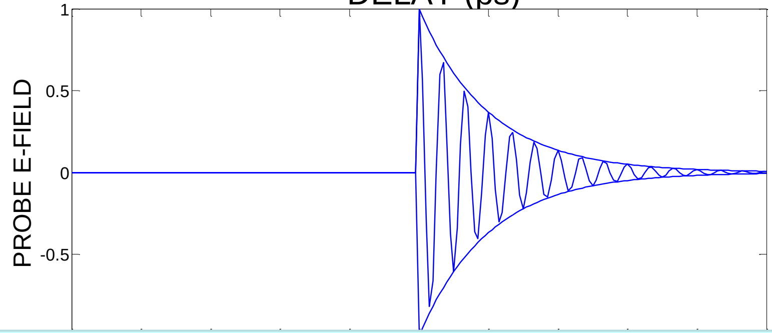
$$W_{JJ'}^{(k)}=X_k\,\gamma_k^2\left(N_{J'}^{(k)}-N_J^{(k)}\right)b_{JJ'}\,F_J^{(k)}$$

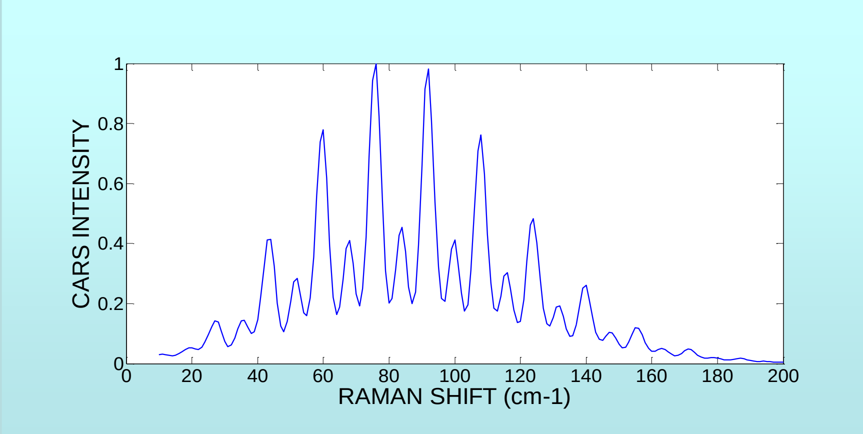
- This function, χ, is then multiplied by the *measured* probe electric field to obtain the CARS signal field

$$E_{CARS}(t-\tau)=E_{pr}(t-\tau)\exp\left[i\left(\omega_o(t-\tau)-\phi\right)\right]\times\chi(t)$$

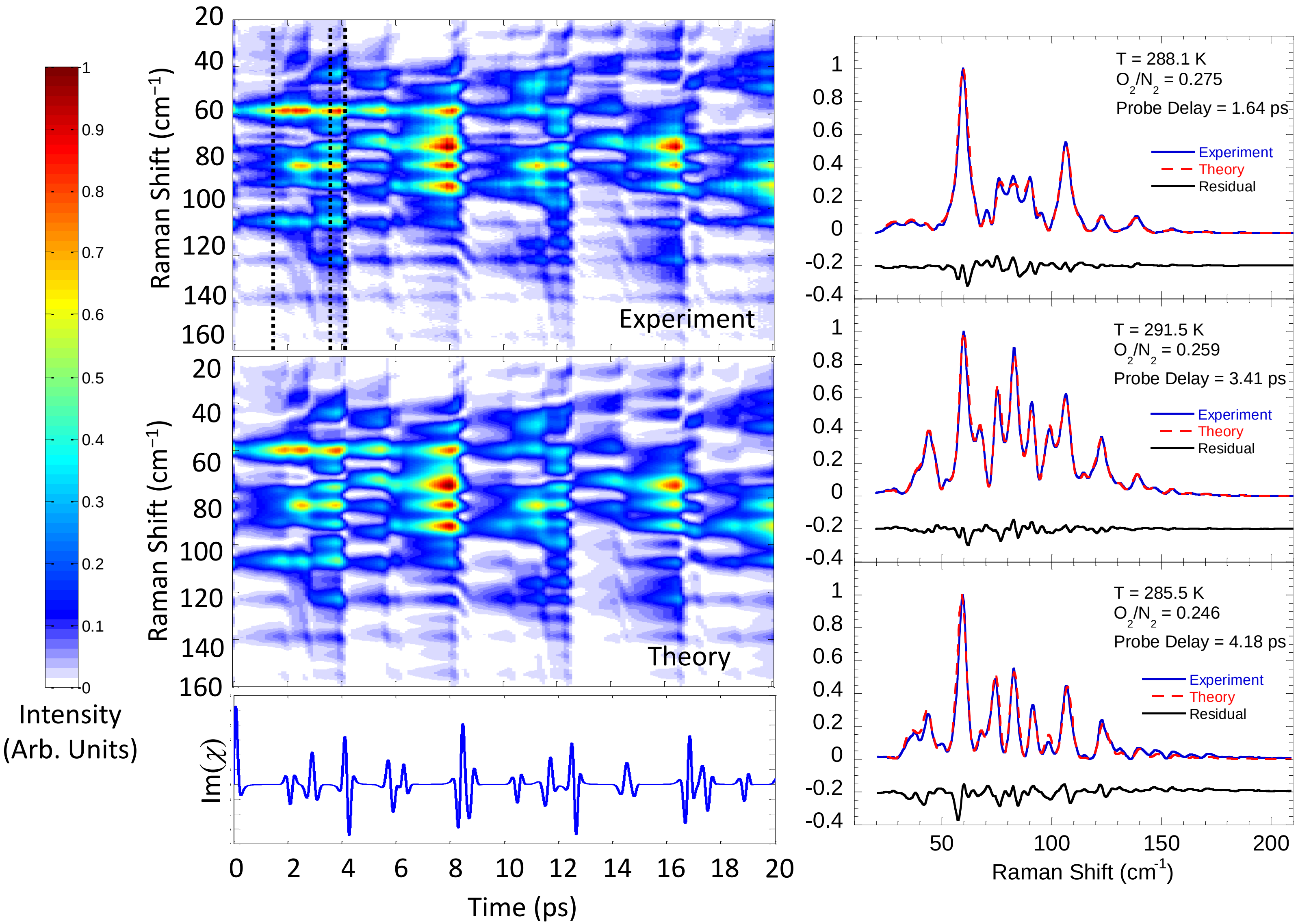
- The CARS spectrum is arrived at by Fourier transform of E_{CARS}
- 1-2 spectra/min desktop
- 1500 spectra/hr parallel (250 nodes



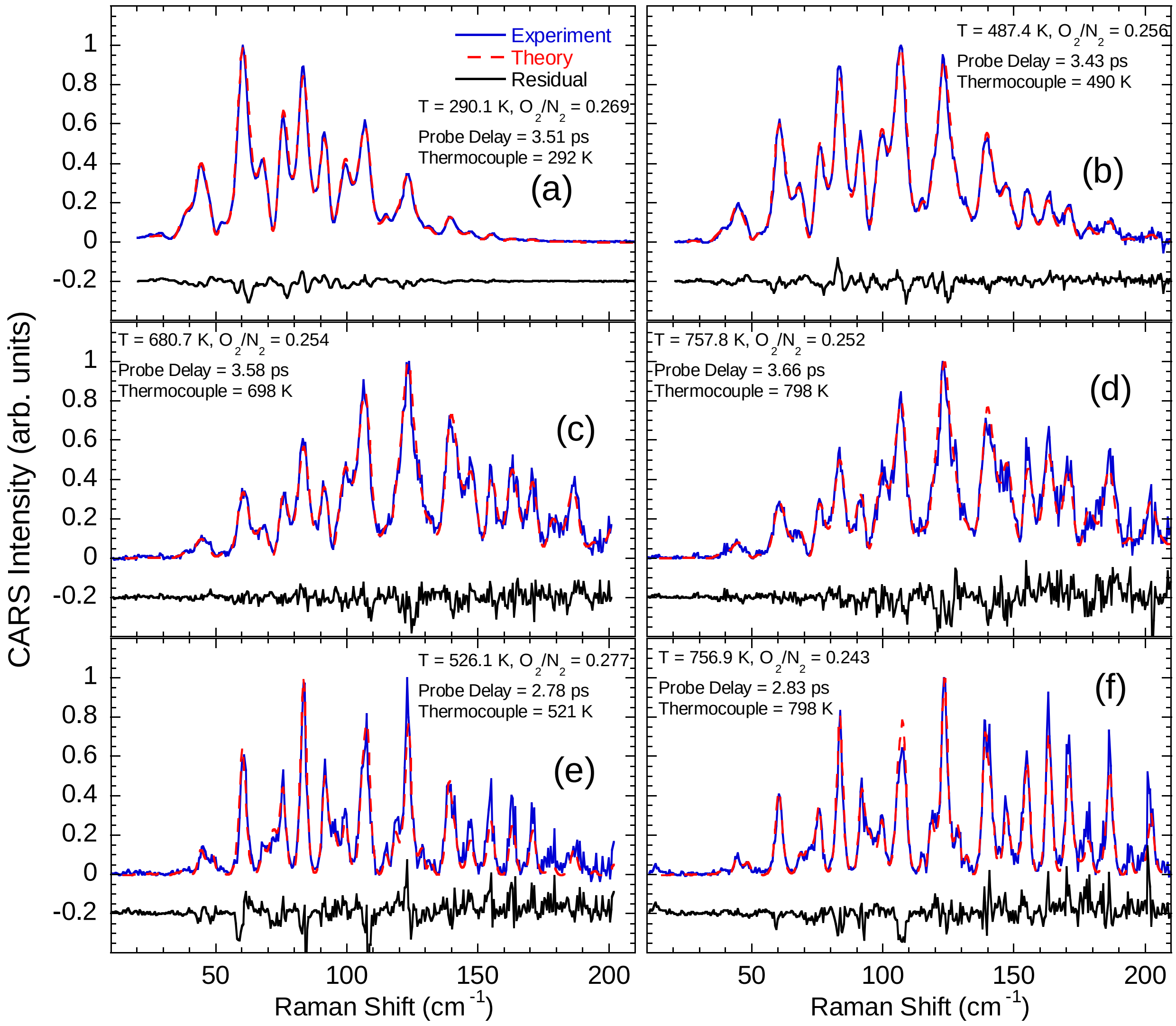




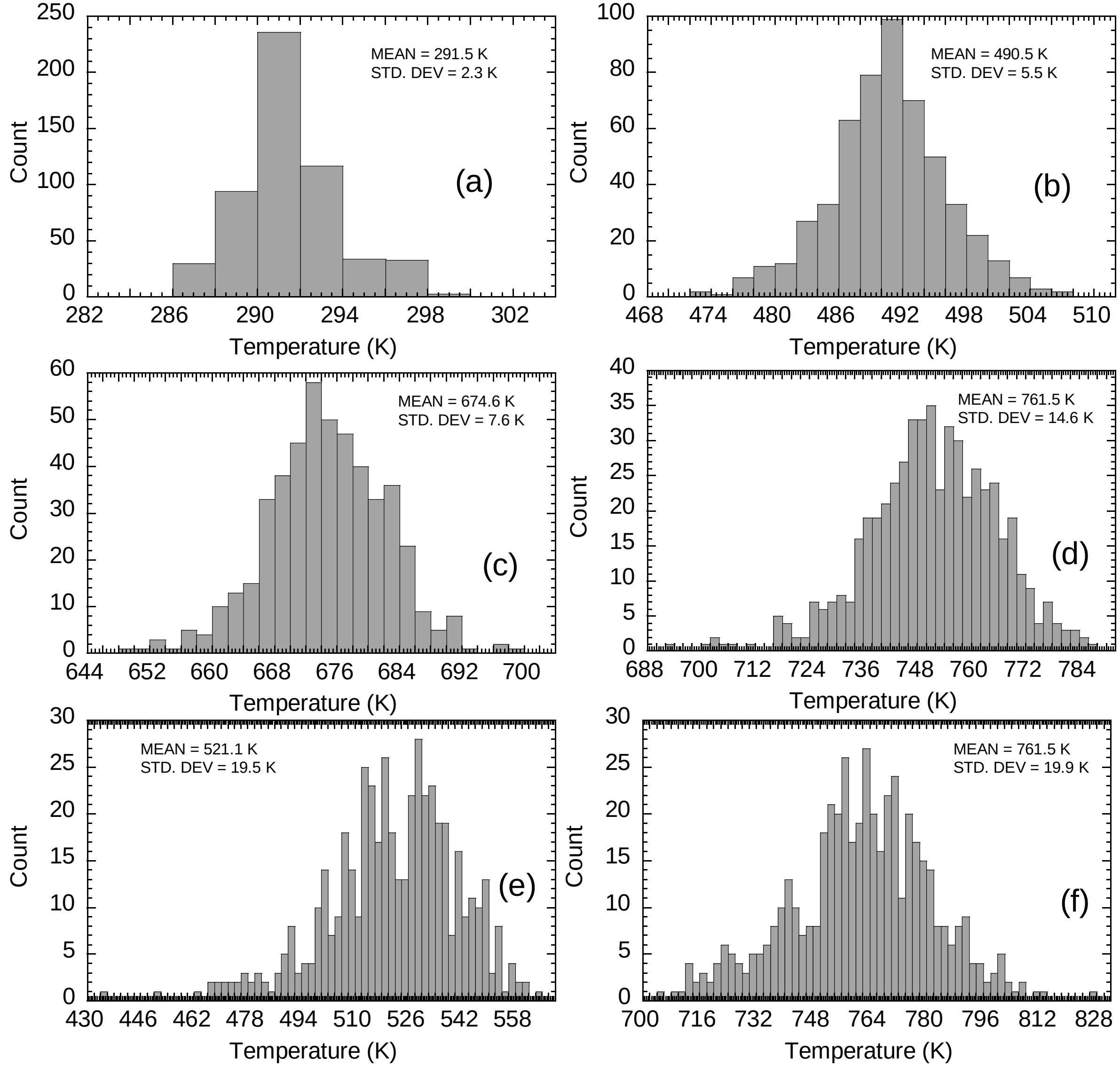
Model validation in room-temperature air: 1.5-ps probe



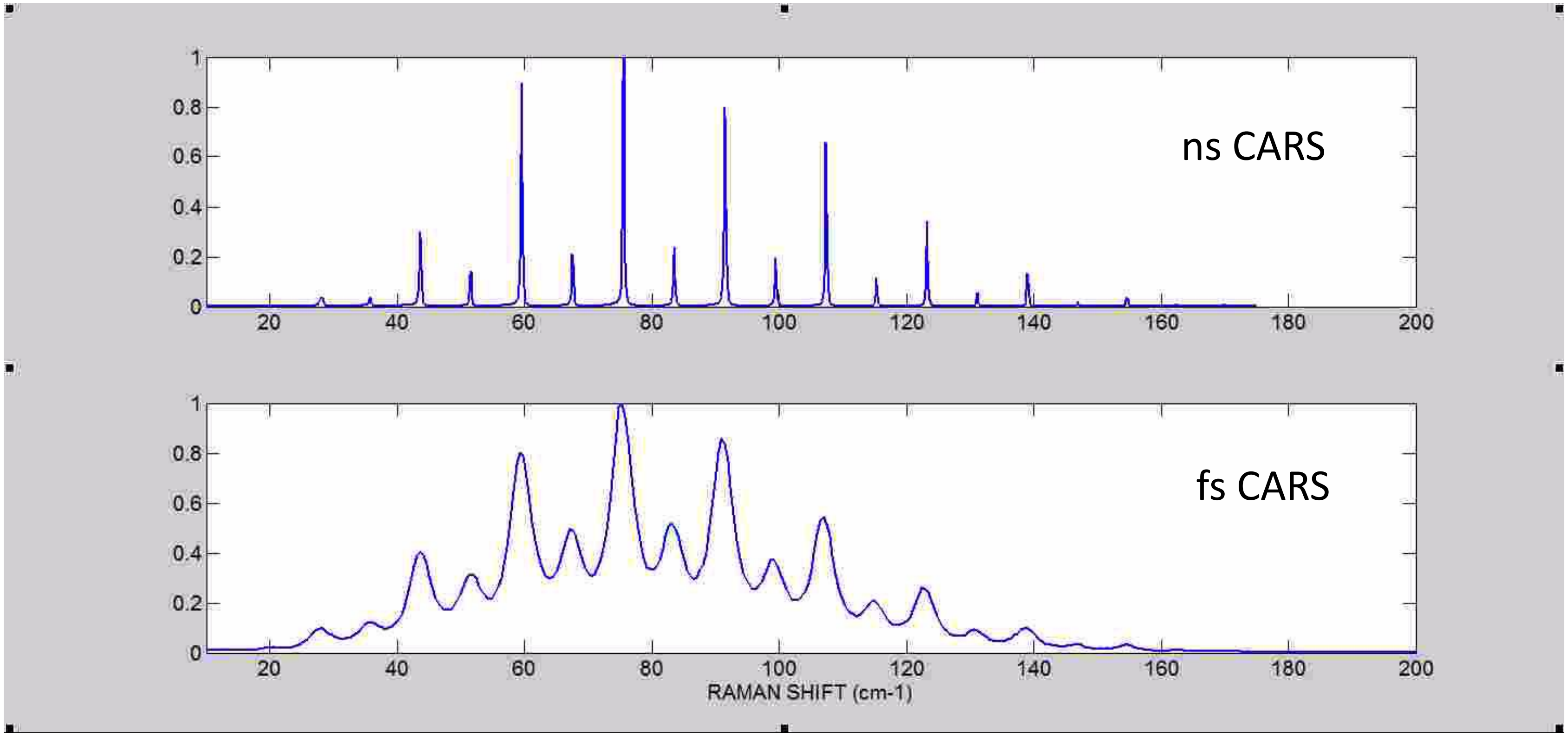
Single-laser-shot spectra



Single-shot temperature histograms



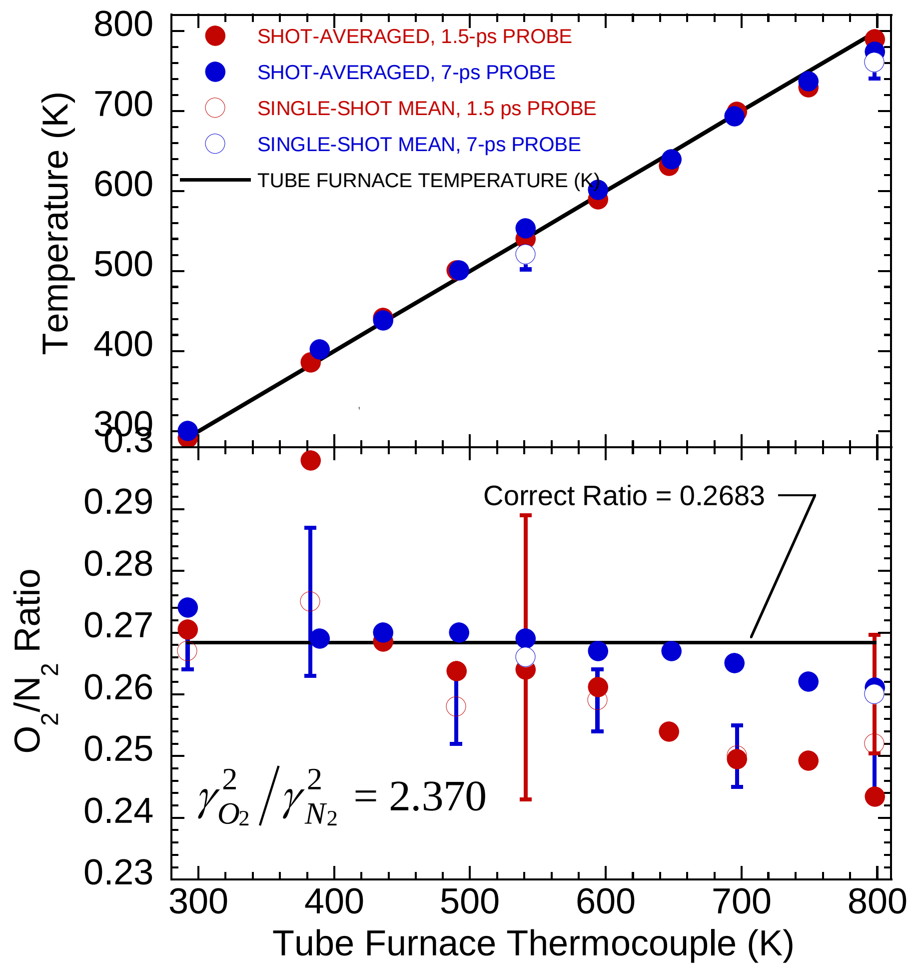
Low noise fs preparation pulses result can result in higher single-laser-shot precision



Room-temperature N_2 spectra

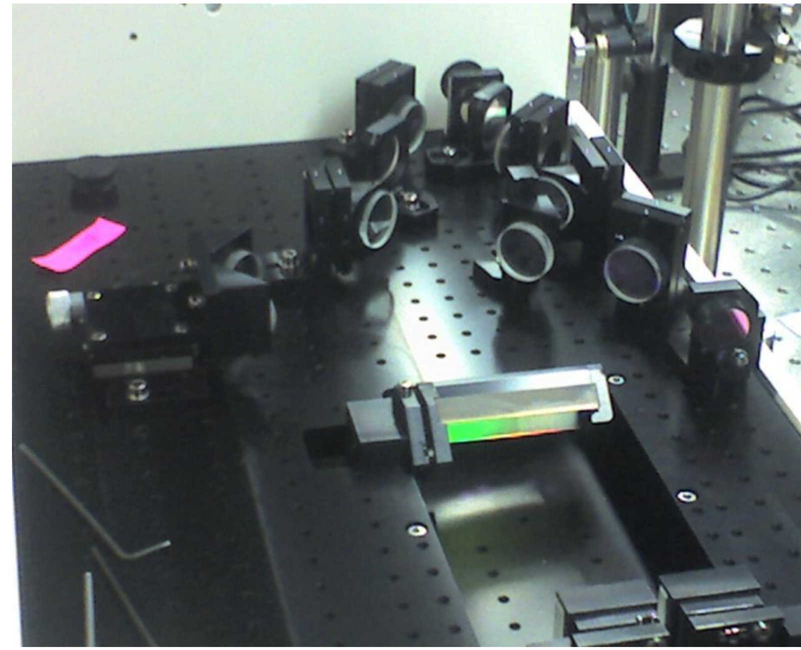
Thermometry results and observations

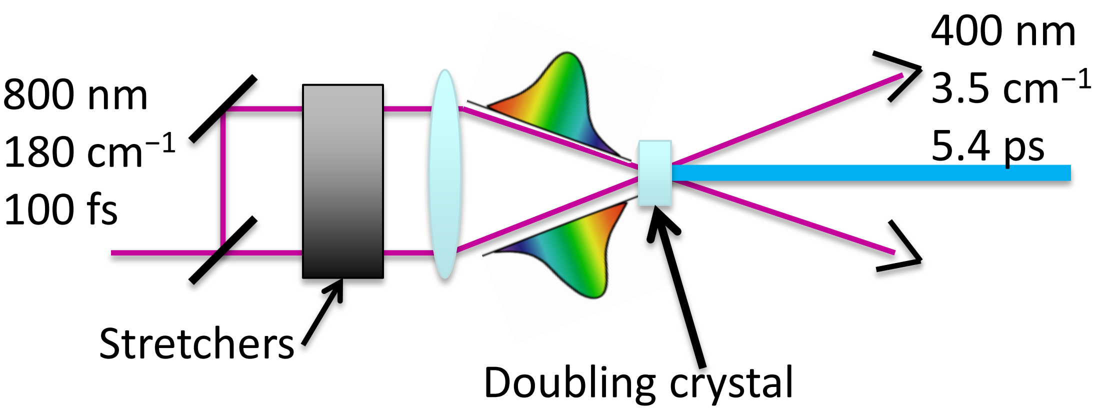
- We have demonstrated single-shot thermometry and concentration measurements in air at temperatures up to 800 K
- Air temperatures are generally within 2.5% of the tube-furnace control thermocouple, with some exceptions at T = 800 K.
- Single-shot temperature precision is 1-2%. This superior to all but the best results obtained with ns-CARS
- Excellent shot-to-shot spectral repeatability from low-noise fs preparation pulses
- O2 measurements need additional refinement
- Probe pulse energy must be increased to reach flame temperatures

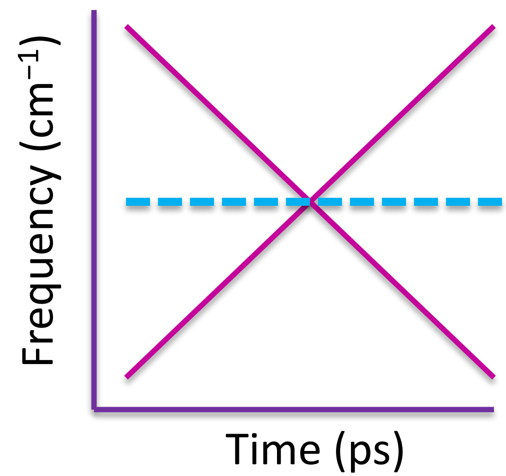


SHBC for Efficient Probe Generation

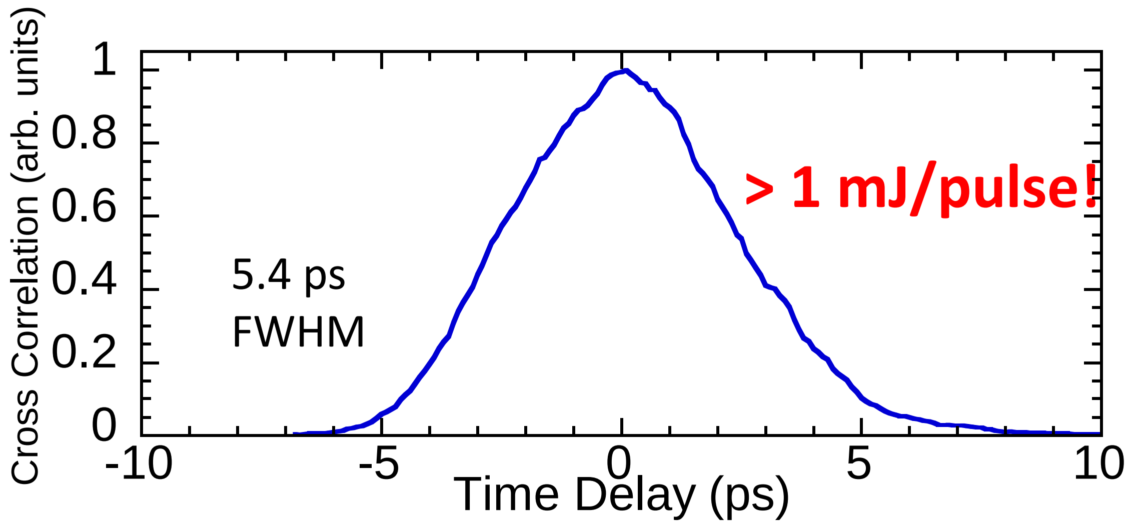
- SHBC = Second Harmonic Bandwidth Compressor (Spectra Physics/Light Conversion)
- Enabling technology
- This device is 35% or more efficient
- It has increased probe pulse energy from 10 μJ to as much as 1.1 mJ
- Output bandwidth is ~3.5 cm^{-1} (~5.4 ps)
- New probe (and signal) shifted to 400 nm
- Will rapidly extent our present CARS implementation to flame temperatures

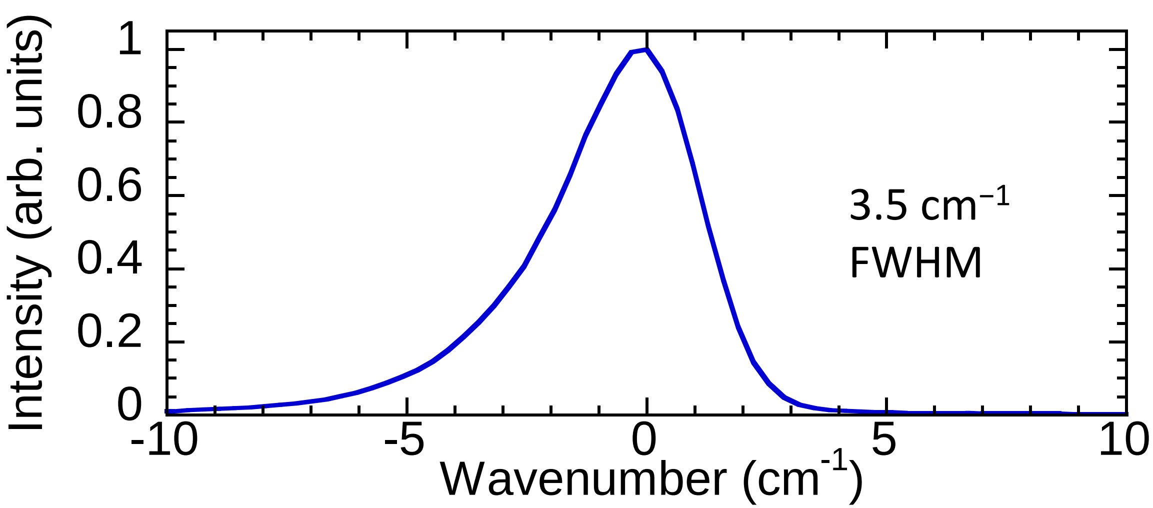




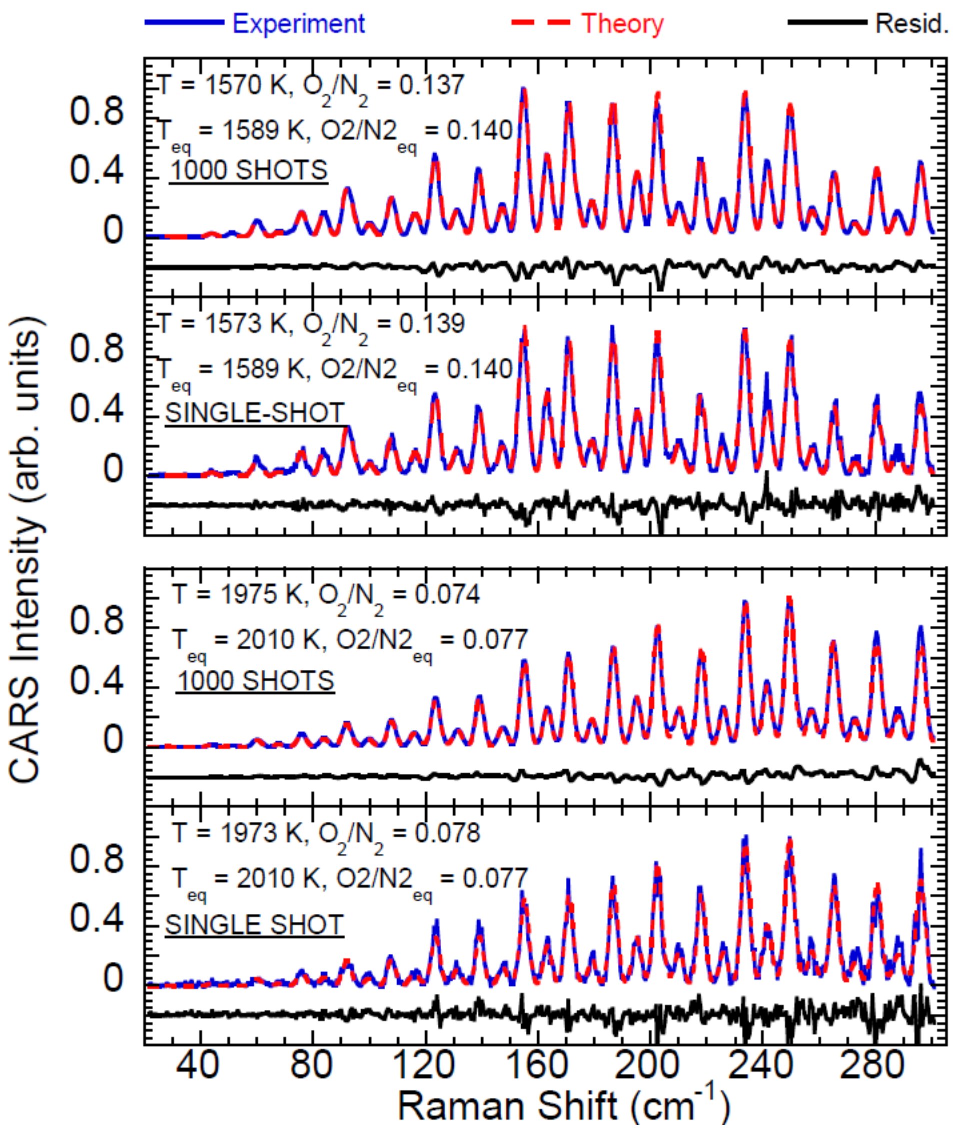


SHBC Output Spectrum and Pulse Shape





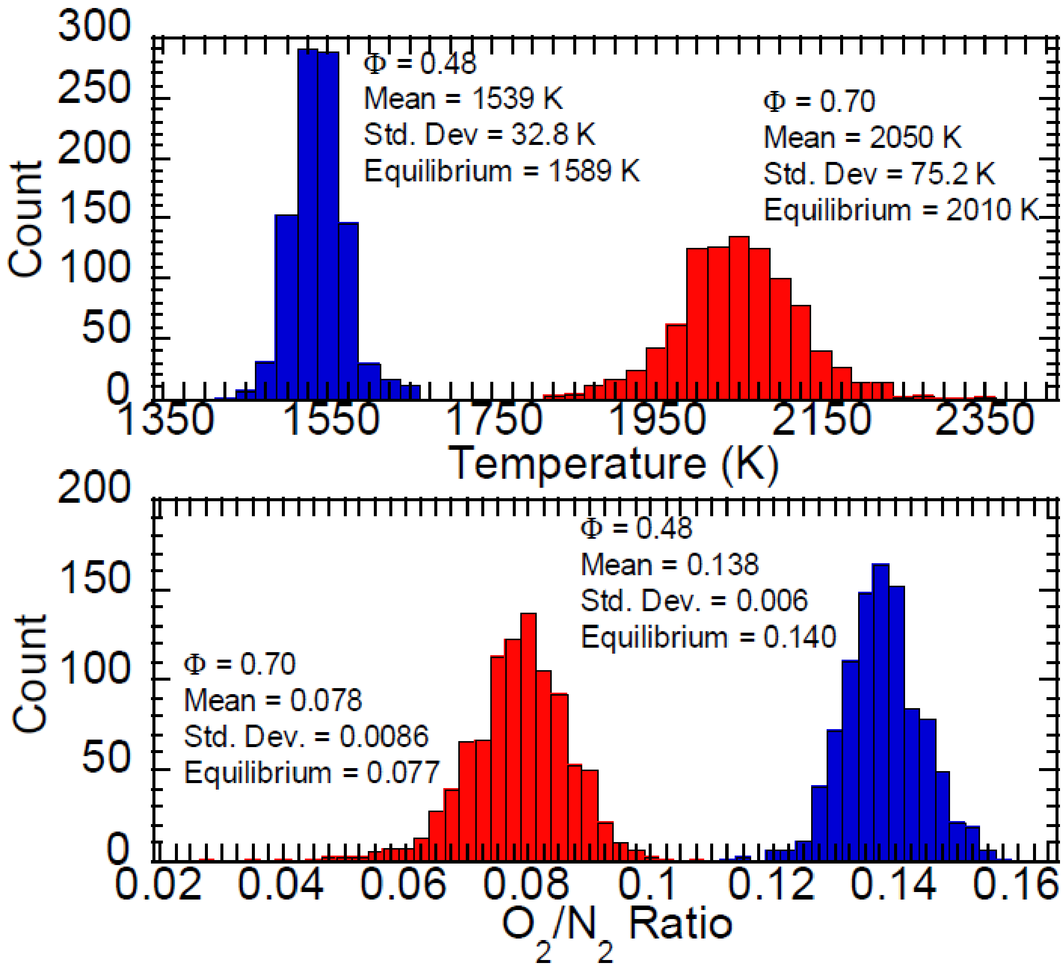
Initial SHBC Rotational CARS Flame Spectra



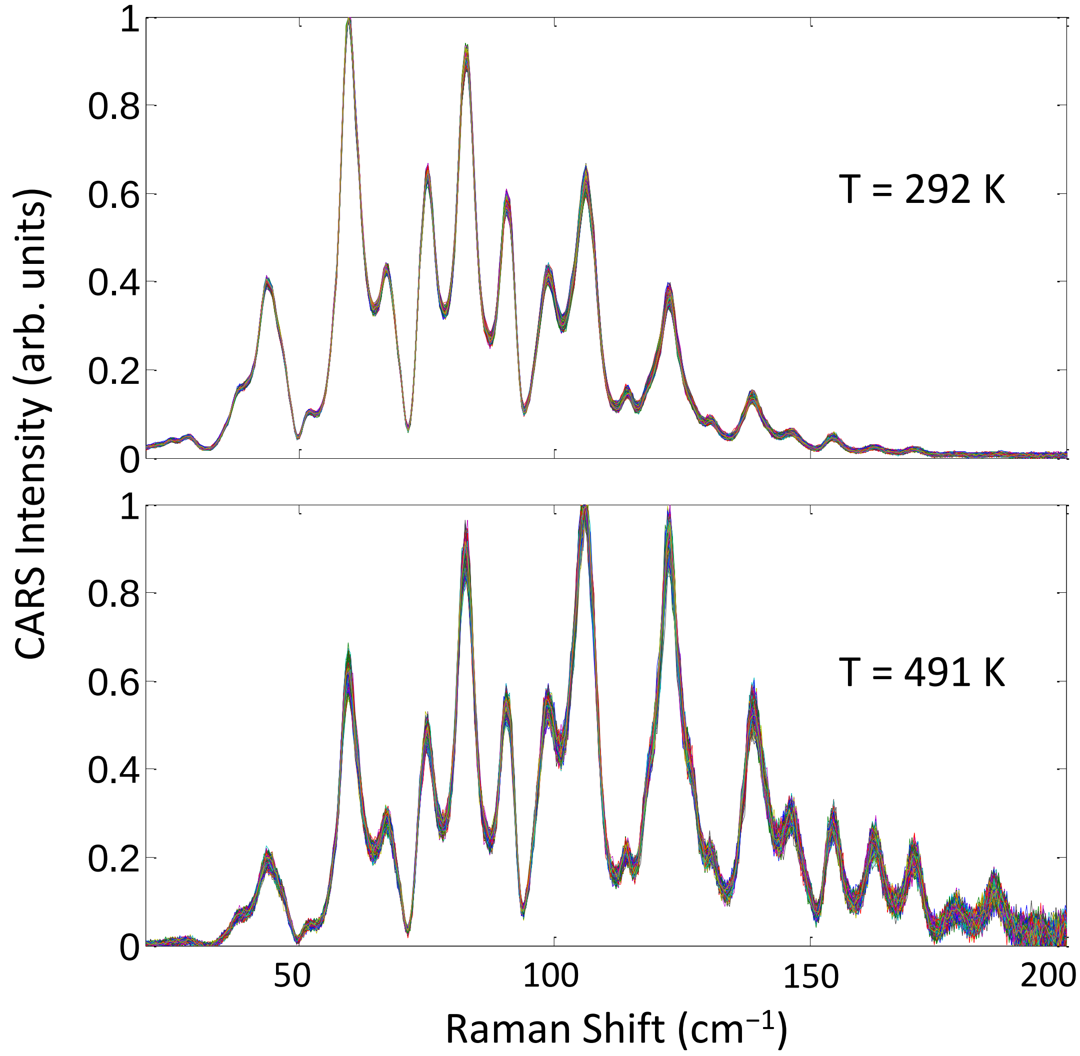
- Near-adiabatic H_2/air flame stabilized on a Hencken Burner
- Temperature and O_2/N_2 ratio estimated from adiabatic equilibrium
- 100-fs pump and Stokes beams at 800 nm
- $\varepsilon_{pump} = \varepsilon_{stokes} = 50\mu J$
- $\varepsilon_{probe} = 1000\ \mu J$ at 400 nm
- High probe energy and ω^2 scaling of CARS signal enable flame measurements
- Single-shot data acquired at 1 kHz for temperatures in excess of 2000 K!

Single-Laser-Shot Results From a Flame

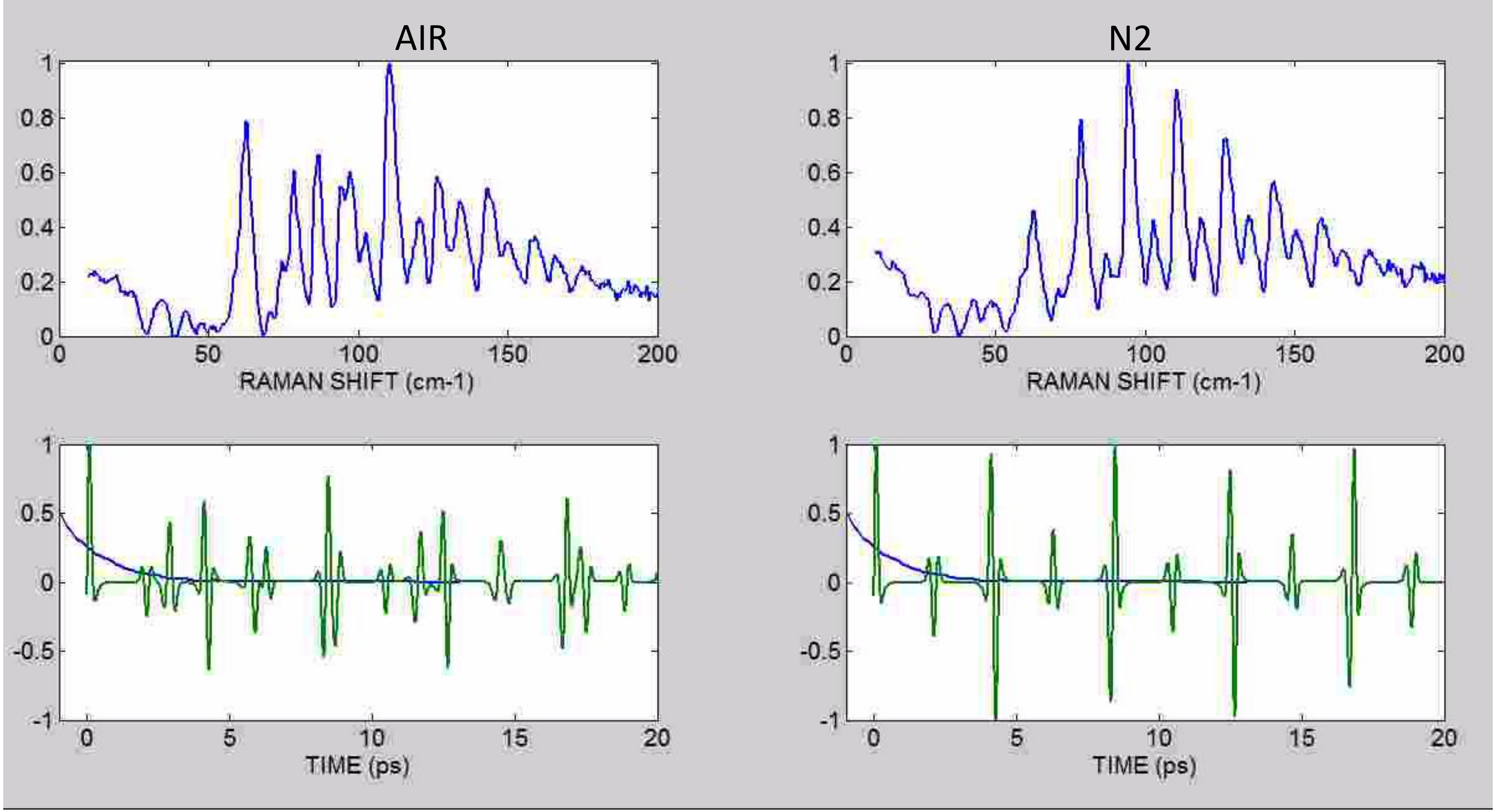
- Single-laser-shot data at $\Phi = 0.48$ and 0.70 acquired at 1 kHz
- Good signal-to-noise with EM gain
- 1000 shots fit to model here
- Temperature measurement precision 2.1% and 3.7%, respectively
- ns R-CARS is typically 3-4%
- O_2/N_2 precision is 4.2% and 11%, respectively
- Good agreement with adiabatic equilibrium
- Temperature to within 3% of equilibrium
- O_2/N_2 within < 1%
- Well within uncertainty due to metered flow rates
- Can we achieve 1% precision without EM gain?



1000 Single-Laser-Shot Spectra Show Superior Repeatability



Unsteady CARS spectra from room-temperature gas: single-etalon probe

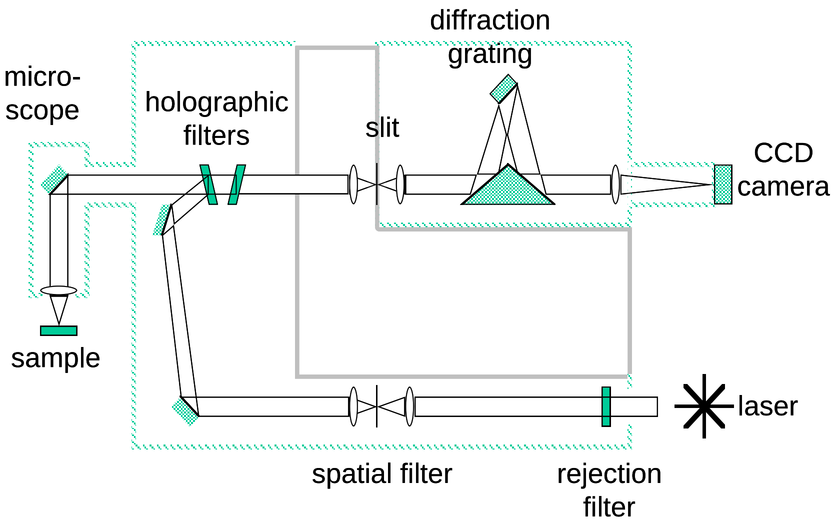


Summary of Single-Laser-Shot Results

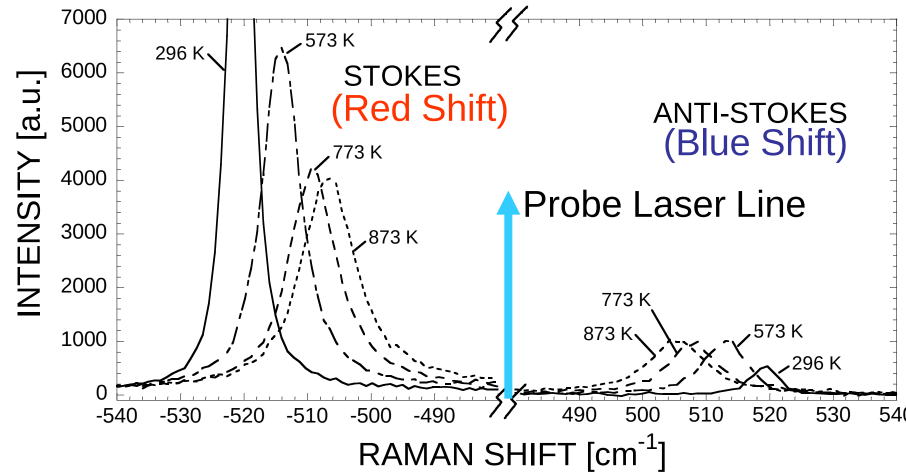
Probe Duration (ps)	Peak counts	Furnace Thermocouple (K)	Mean CARS Temp. (K)	RMS CARS Temp. (K / %)	Mean CARS O_2/N_2	RMS CARS O_2/N_2 (-- / %)
1.5	38,300	292.0	291.5	2.3 / 0.8%	0.267	0.003 / 1.1%
1.5	9,700	382.4	390.0	6.11 / 1.6%	0.275	0.012 / 4.3%
1.5	3,500	490.0	490.5	5.5 / 1.1%	0.258	0.006 / 2.5%
1.5	6,500*	594.2	582.9	6.2 / 1.1%	0.259	0.005 / 1.9%
1.5	49,100*	696.7	674.6	7.6/ 1.1%	0.250	0.005 / 2.0%
1.5	27,000*	798.0	751.1	14.6 / 1.9%	0.252	0.0082 / 3.2%
7.0	3,300*	540.9	521.1	19.5 / 3.7	0.266	0.023 / 8.6
7.0	34,000*	798.0	761.5	19.9 / 2.6	0.260	0.0096 / 3.9

* Indicates that detector's electron-multiplying gain was used.

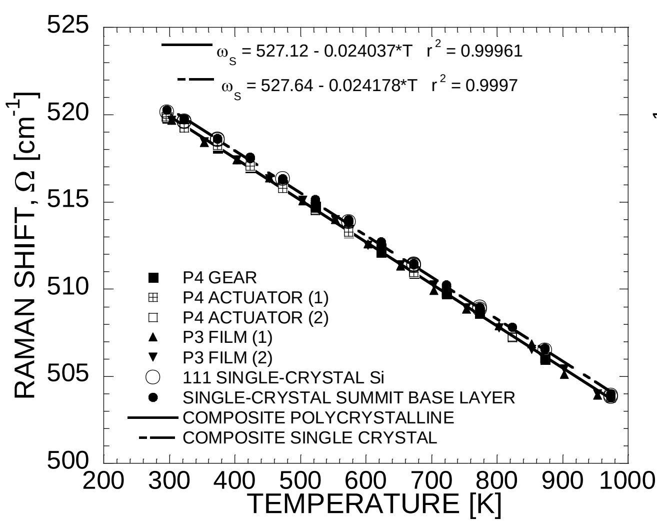
Micro-Raman Diagnostics for MEMS Investigations

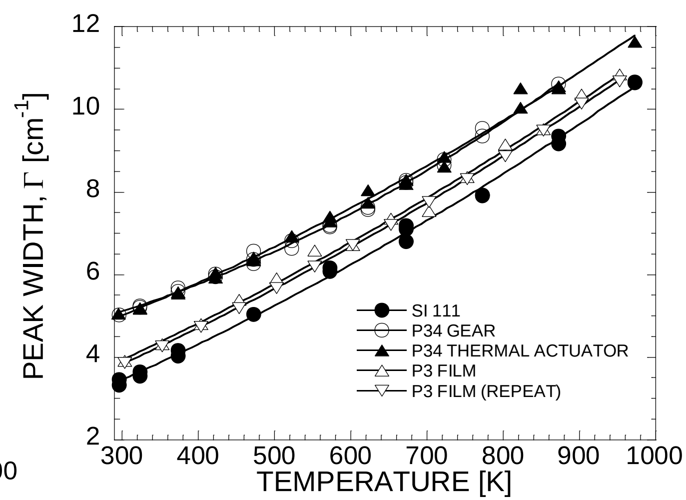


Schematic of Raman Microscope System



Raman Spectrum of Silicon at Various Temperatures

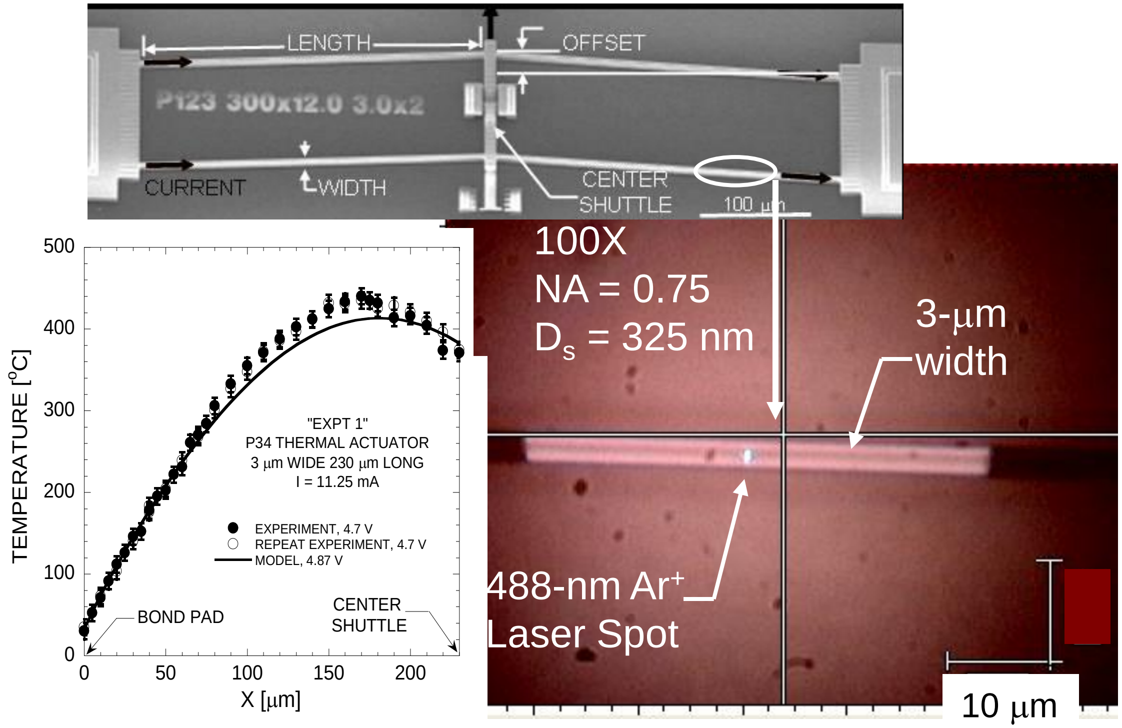


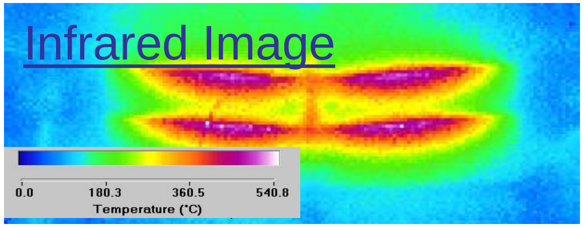


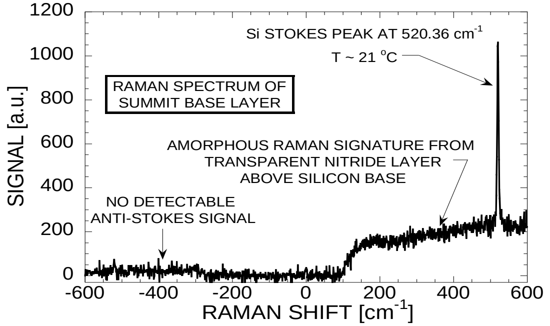
Raman Calibration Curves for Si

Application Example – Thermal Actuators

- Chevron Style Thermal Actuators
- Electric Current = Device Motion
- Potential Applications Include
    - Nonvolatile Memory
    - Mechanical Switches
    - Driving of Other Microsystems
- Small 3-μm leg widths make spatial resolution a challenge
- First-ever resolved temperature profiles obtained from working thermal actuators
- Dramatic improvement over existing infrared capability
- Results have been used for validation of device heat-transfer models



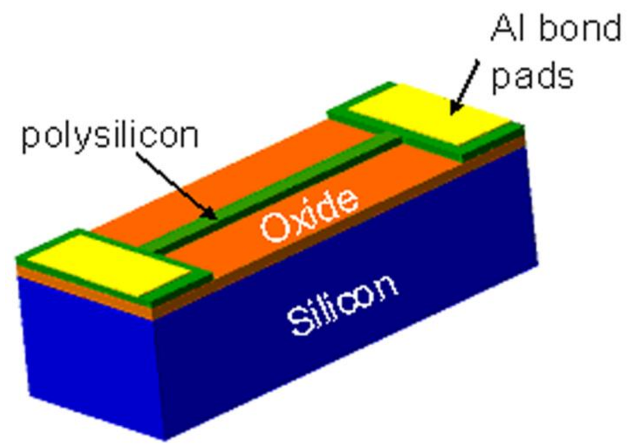




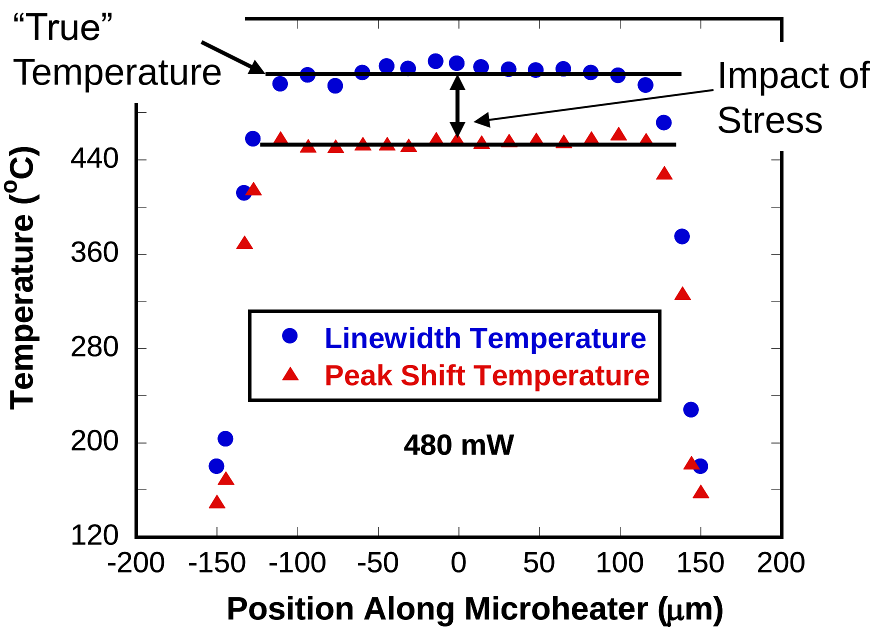
Kearney *et al.*, *J. MEMS* **15**, 314-321, 2005.

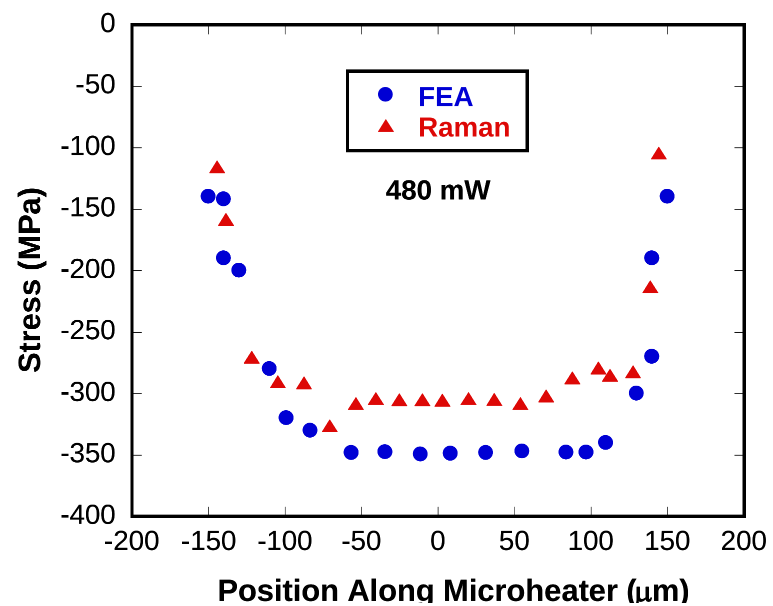
Multi-Parameter Capability -- Simultaneous Temperature/Stress Mapping

- Raman peak position is further influenced by stress in the material
- Raman peak width is not impacted by stress
- Difference between “peak” and “linewidth” temperatures is used to determine the avg. stress in a Si microbeam



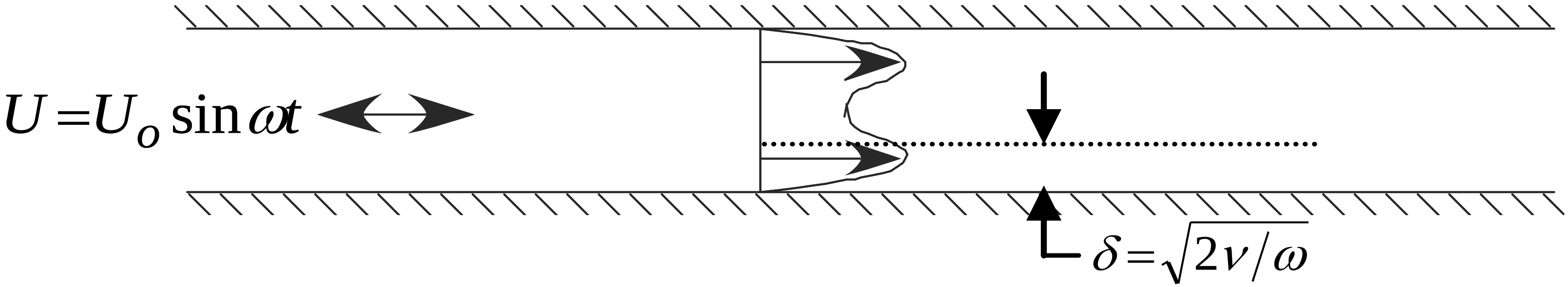
Unreleased Polysilicon Test Beam





Beecham, Graham, Kearney *et al.*, *Rev. Sci. Inst.* **78,** 061301, 2007 (invited).

PIV Investigation of Unsteady Turbulent Channel Flow

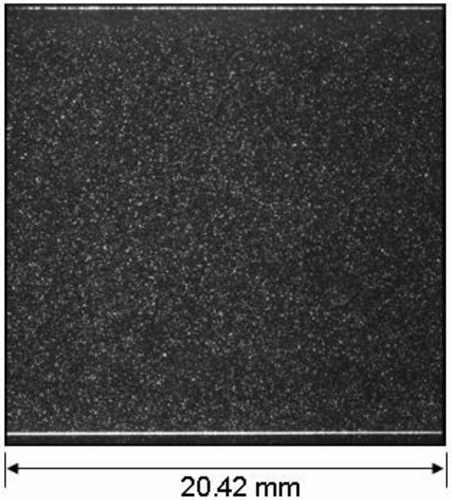


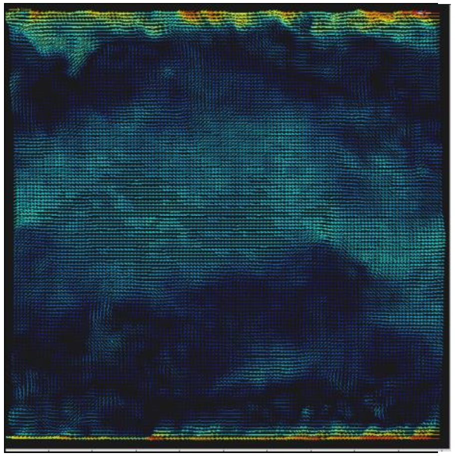
- "Pulsating" Flow → Non-zero time-mean flow "AC" and "DC"
- "Oscillating" Flow → Zero time-mean flow, "AC" only
- When the AC component dominates there are 4 flow regimes parameterized by $Re_\delta = U_o\delta/\nu$
  - Laminar: $Re_\delta < 100$
  - Disturbed Laminar: $100 < Re_\delta < 500$-550
  - Intermittently Turbulent (IT): $500 < Re_\delta < 3000$
  - Fully Turbulent: $Re_\delta > 3000$

II. Dual-Magnification PIV Required for Wall Shear Stress and Bulk Flow Characterization

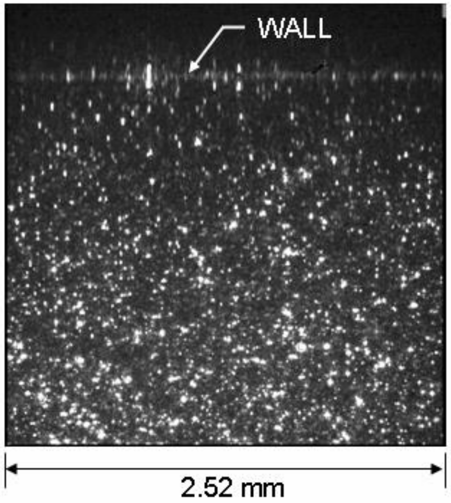
- Low Magnification (0.33:1)
  - Images full channel
  - Characterize bulk flow
- High Magnification (3.45:1)
  - Images near-wall region
  - Measure wall shear stress
- DaVis Commercial Software
  - Iterative cross-correlation PIV interrogation
  - 64×64 window coarse grid used to estimate velocity magnitude <u>and direction</u>
  - Final result on 16×16 grid

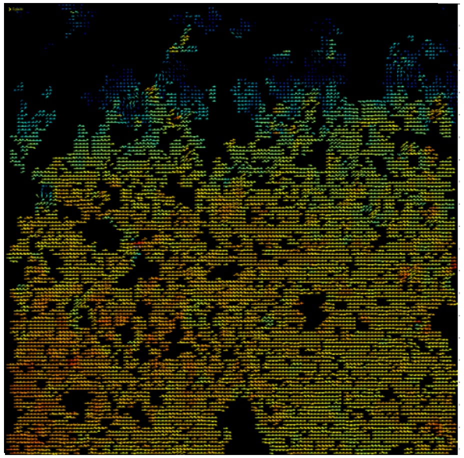
LOW MAGNIFICATION





HIGH MAGNIFICATION

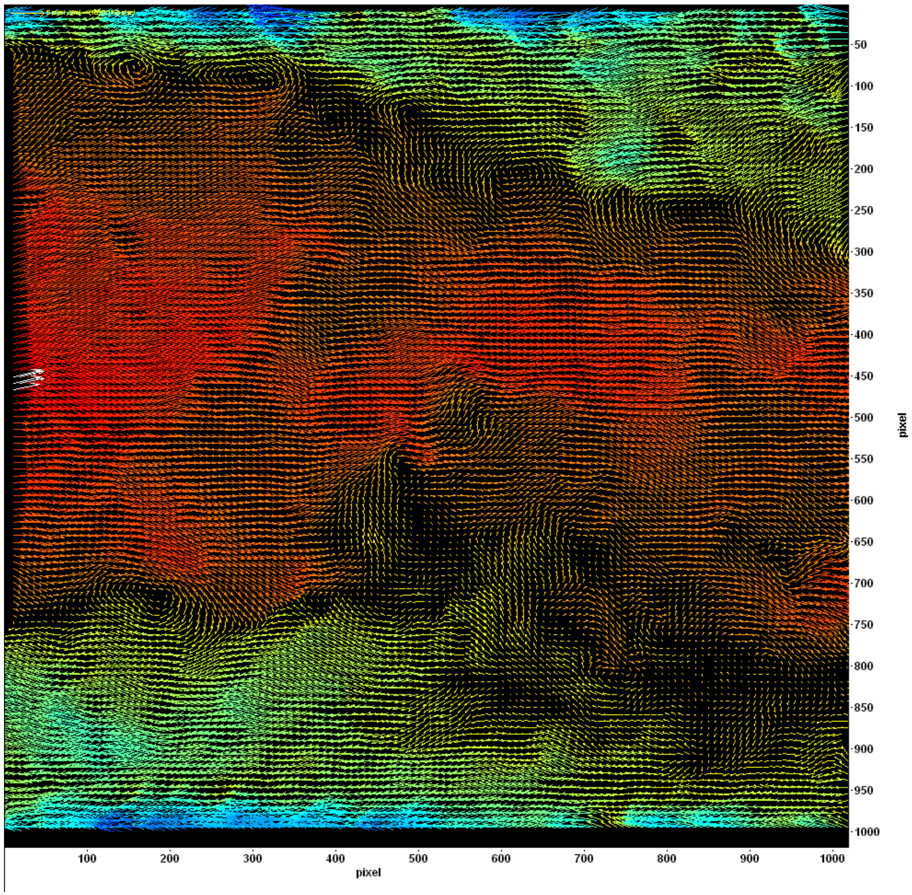


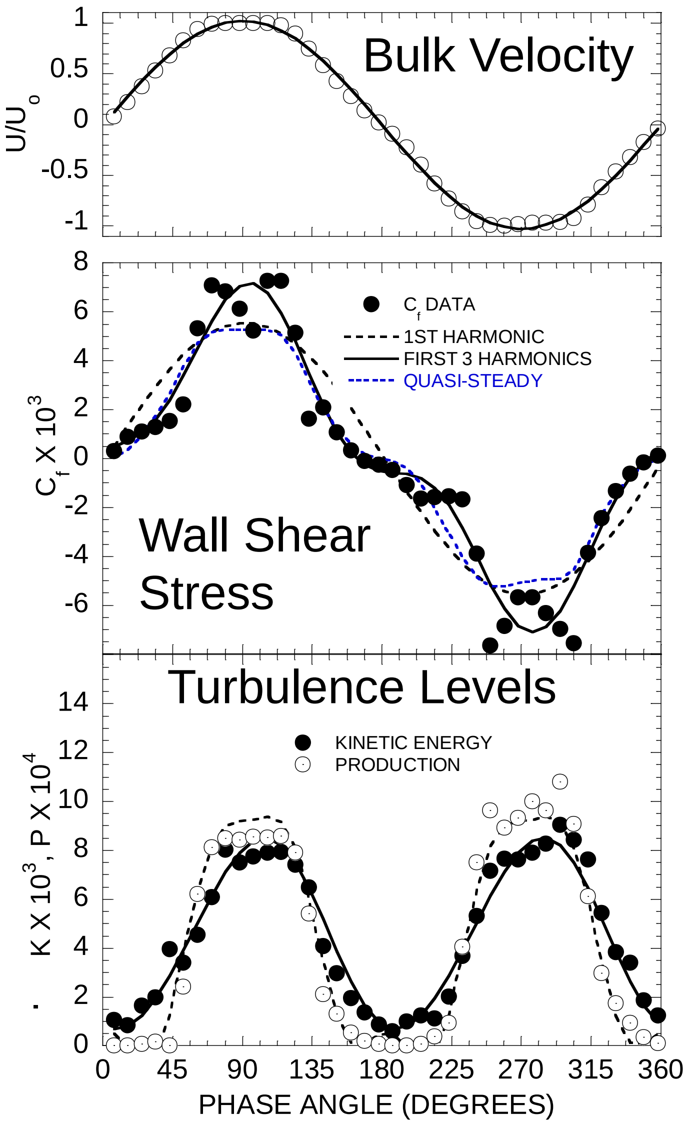


<u>Spatial Resolution in Wall Units</u>

τ (Pa)	Low Mag. 160 μm res.	High Mag. 19.6 μm res.
0.1	1.7	0.2
6	13.1	1.6

Test Case, SEAWOLF Channel Flow at Re_δ = 2033





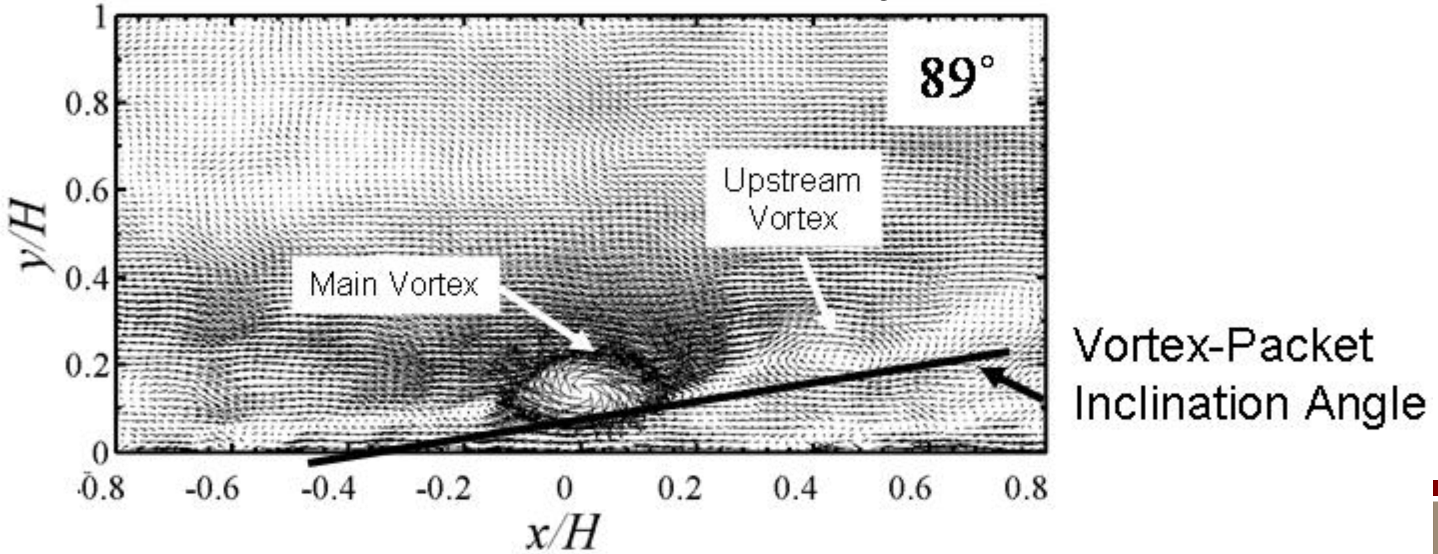
Conditional Averages Can Confirm Hairpin-Vortex Signatures are/are not a Persistent Feature in Oscillating Wall Flows

Sandia National Laboratories

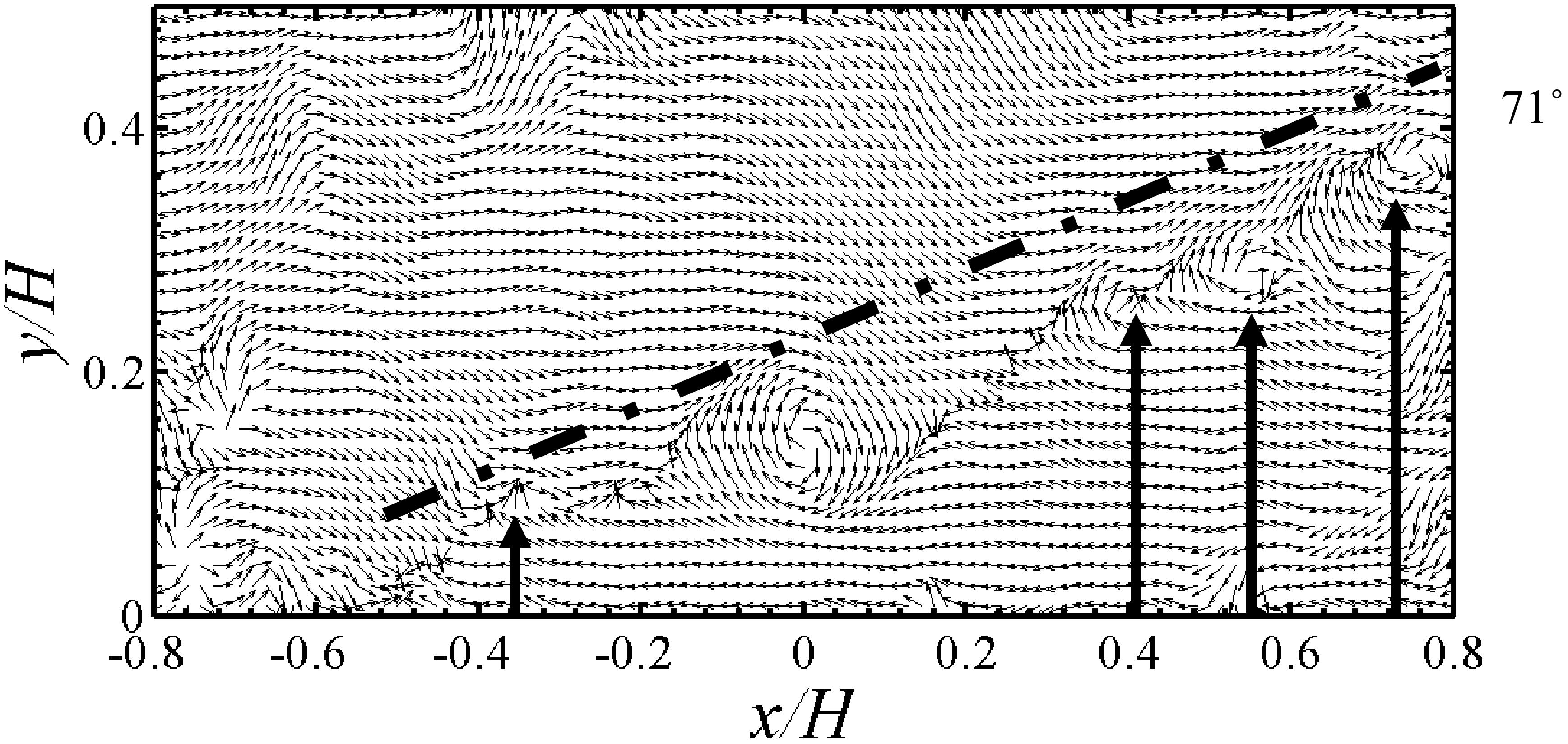
- We can further ask the question, "what does the velocity field look like given a vortex near the wall?"
- The answer is arrived at by estimating the conditional average, as performed on steady flow data by Christensen and Adrian (JFM, 2001)

$$\left\langle u_j(x+r,y) \middle| \lambda_{ci}(x,y_o) \right\rangle \approx \frac{\left\langle \lambda_{ci}(x,y_o) u_j(x+r,y) \right\rangle}{\left\langle \lambda_{ci}(x,y_o) \lambda_{ci}(x,y_o) \right\rangle} \lambda_{ci}(x,y_o)$$

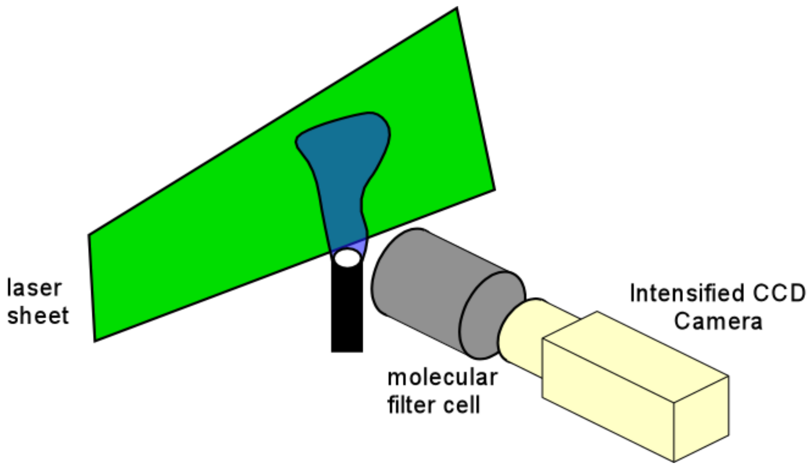
- The average velocity field **given** a swirling strength event, λ_{ci}, at distance y_o from the wall
- Performed on our phase-resolved data sets for $y_o/H = 0.14$ ($45 < y^+ < 120$)



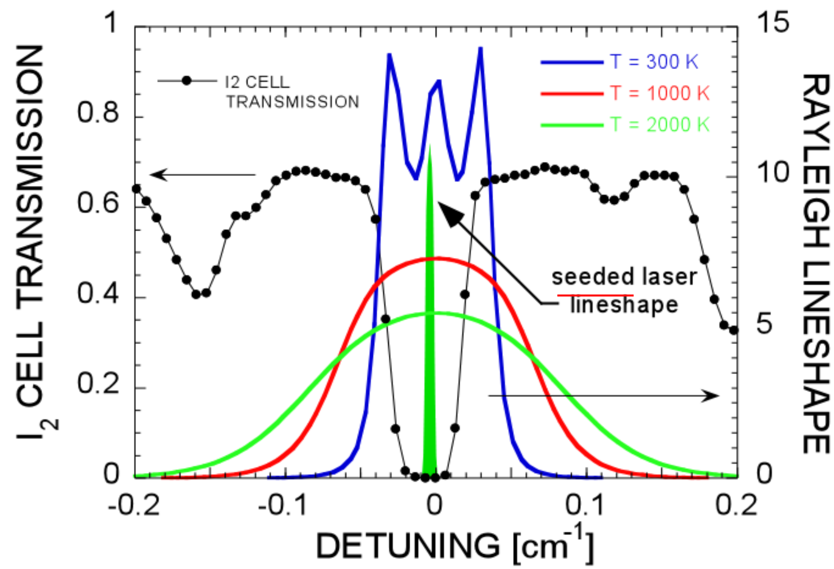
Conditional Velocity Field – Late Acceleration Stage



Filtered Rayleigh Scattering (FRS) for Combustion Thermometry



Laser sheet imaging through iodine filter



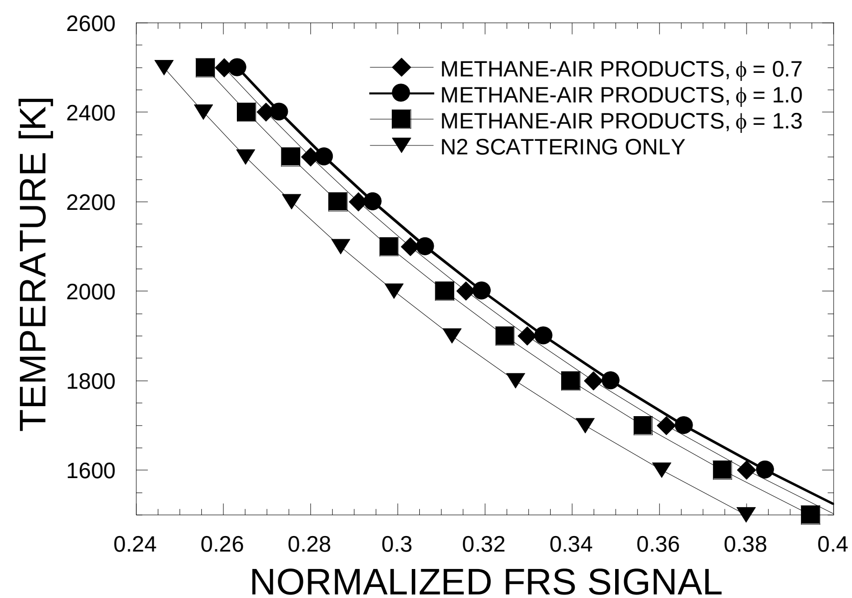
$$S(T,\underline{\chi}) = C\, I_o\, N \sum_k \chi_k \left(\frac{\partial\sigma}{\partial\Omega}\right)_k \int_{\Delta\omega} \Re_k(\omega; T, M_k)\,\tau(\omega)\,d\omega$$

Temperature sensitivity – Doppler broadening

Number Density
N ~ 1/T

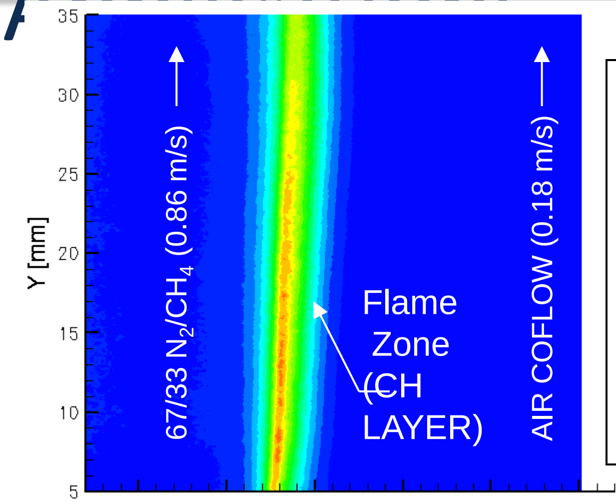
Rayleigh Cross Section
("Scattering Efficiency")

Mole Fraction of Species k

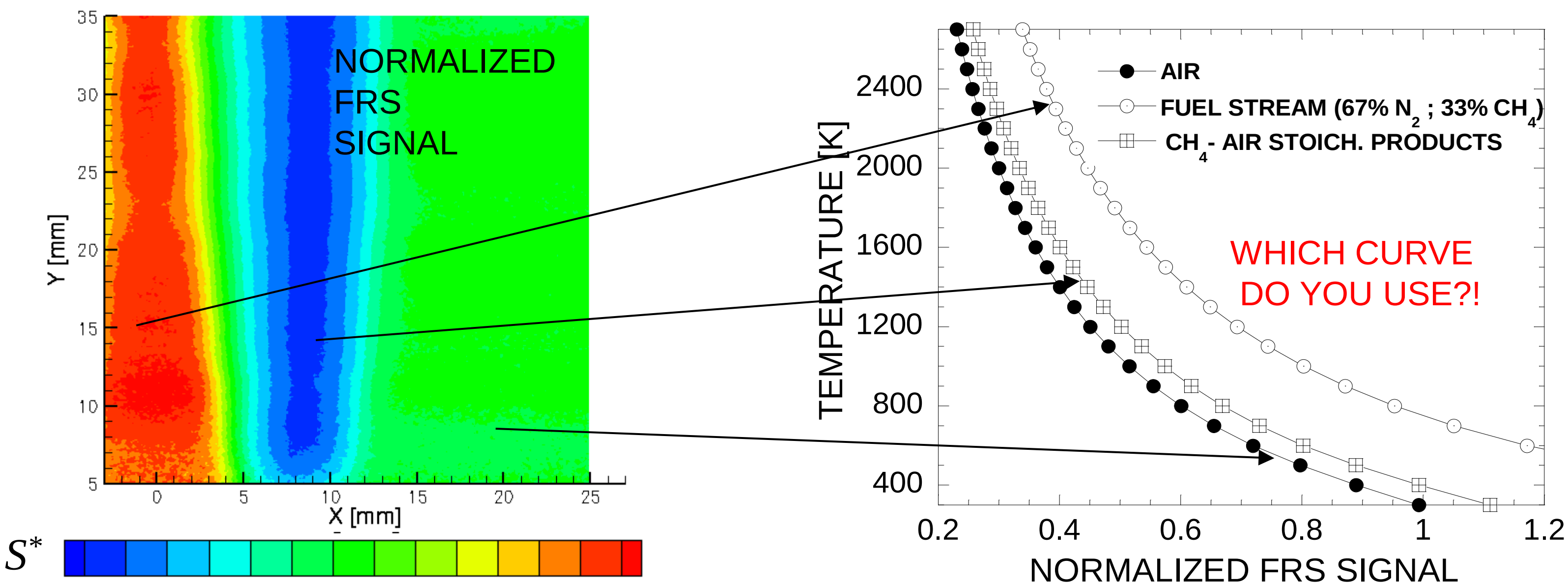


FRS Response is Sensitive to Doppler Shift (Velocity), Temperature and Composition

Composition Bias in Diffusion-Flame Applications



- Large variations in scattering cross section across flame
- Bias errors from 500-1000 K are possible!
- Additional information regarding local chemical composition is required
- Need information on N_2, O_2, CO_2, CO, H_2O, CH_4
- SOLUTION → Raman Imaging of Fuel Number Density in conjunction with a Mixture-Fraction-Based Model to approximate local gas-phase composition

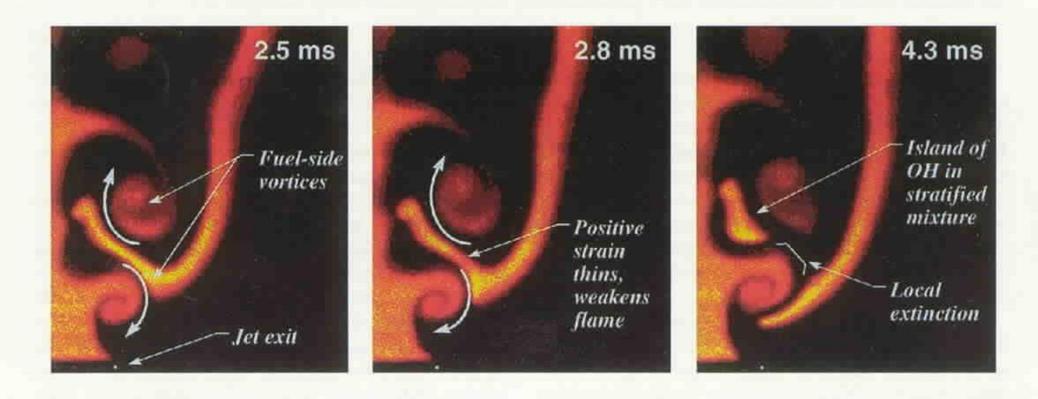


Local Variation in FRS Temperature Response

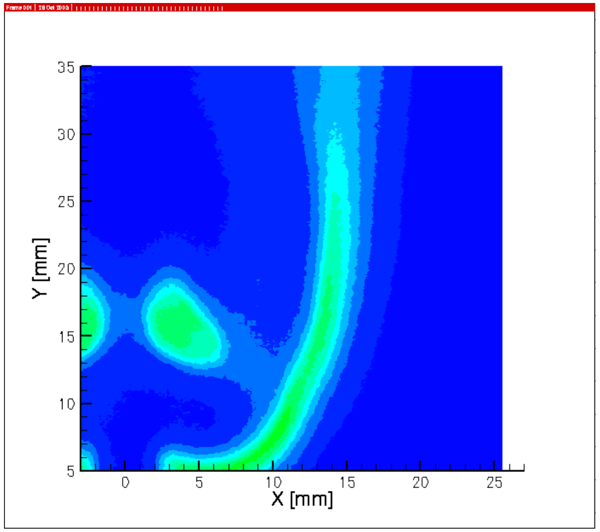
Steady Slot Flame Reference

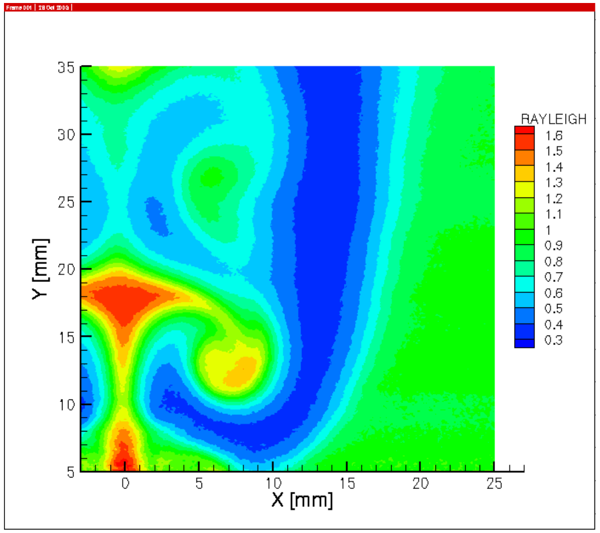
FRS-Raman Probing of Strain Induced Extinction in Flames

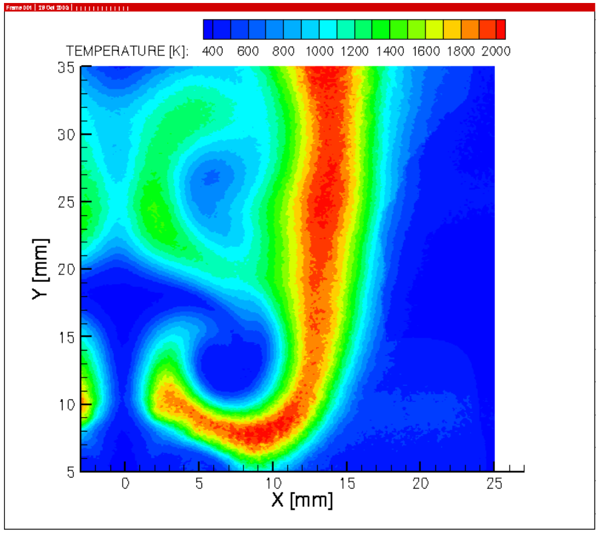
- 2-D slot flame is forced at 90-Hz where 2 consecutive vortices interact with each other and the flame
- Strain is sufficiently high to extinguish the flame
- Facility and diagnostics allow systematic experimental probing of extinction event



Simultaneous OH/Acetone PLIF Flow Viz.
C.J. Mueller and R.W. Schefer, *Proc. Combustion Inst.*, **27**, 1105-1112, 1998







Results – Data Fidelity Single-Shot Measurements from Tube Furnace and Hencken Burner

- Accuracy and precision of N_2 CARS thermometry assessed with 2 lab standards
  - Tube-furnace →thermocouple
  - Adiabatic flat flame → equilibrium calculated temperature
- Ensembles of single-laser-shot CARS spectra acquired for a given furnace/flame setting
- Temperature histograms constructed
  - $T_{CARS,avg} - T_{std}$ = accuracy
  - $T_{CARS,rms}$ = precision
- Single-shots CARS measurements are 14-70 K (3.5 to 5%) precise and 5-40 K accurate for T from 500-2200 K
- Bias at low temperatures results from errors in determining spectral response of detection system and is correctable