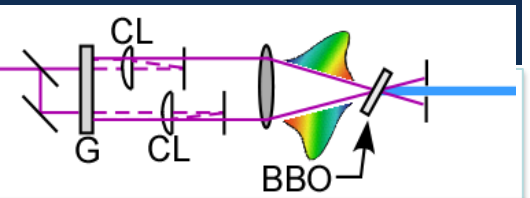
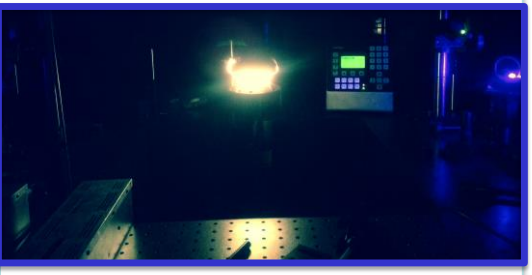
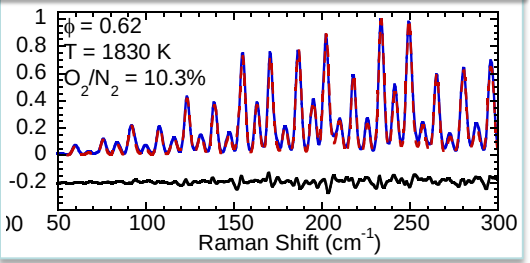
SAND2014-1185C







Femtosecond Laser Raman Combustion Diagnostics

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Sandia National Laboratories

Exceptional service in the national interest

U.S. DEPARTMENT OF ENERGY

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Preview

- Background – 5 mins
  - Laser combustion diagnostics
  - Sandia applications in “hostile” environments
- Coherent anti-Stokes Raman scattering (CARS) – 10 mins
  - Fundamentals with nanosecond laser pulses
  - Application to meter-scale fire measurements
- Ultrafast (fs/ps) CARS development – 30 mins
  - Why ultrafast?
  - Time domain Raman processes and proof-of-concept
  - Game-changing advance: SHBC
  - Flame measurements: accuracy and precision
  - Application to composite fire problem
- New ultrafast applications to shock physics (5 mins)

Laser-based diagnostics empower combustion research

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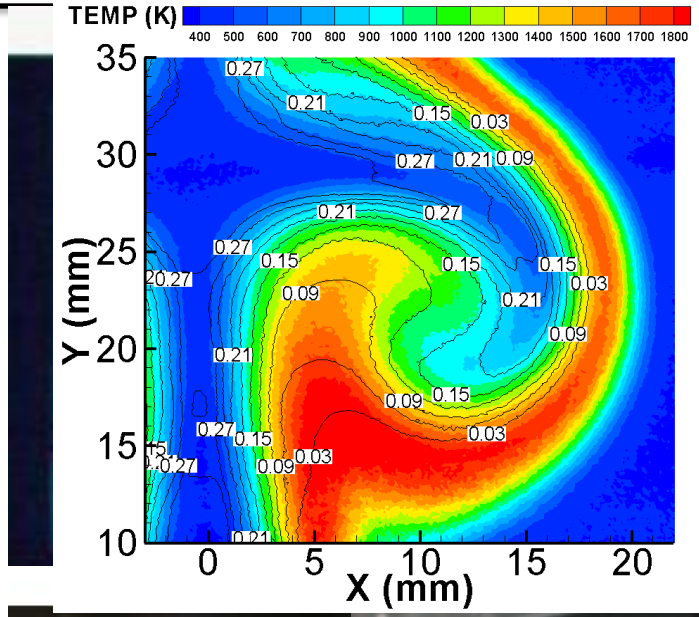
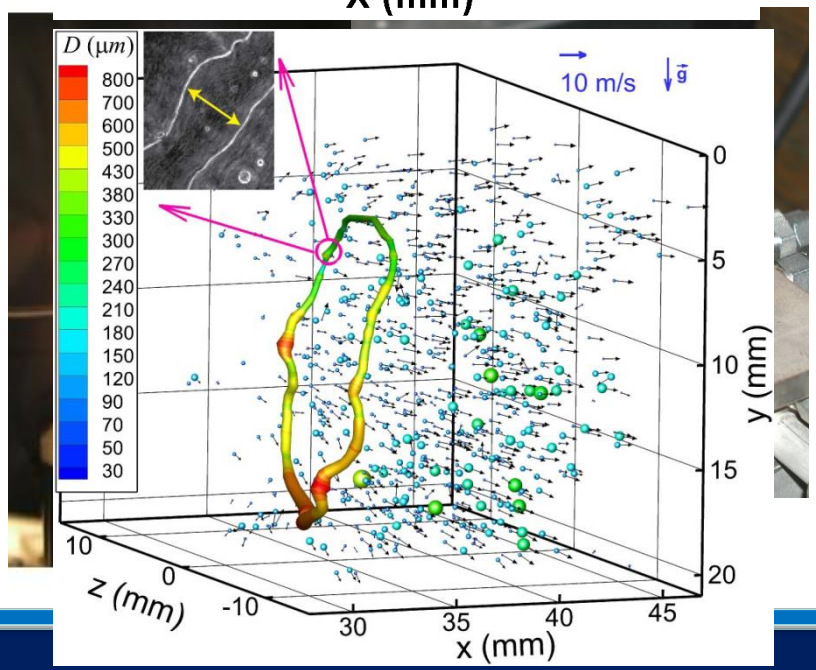
- Non-perturbing
- Free of radiation and insertion errors
- 2-D or even 3-D ***quantitative*** imaging
- Multiple parameters (T/species/soot/velocity…)

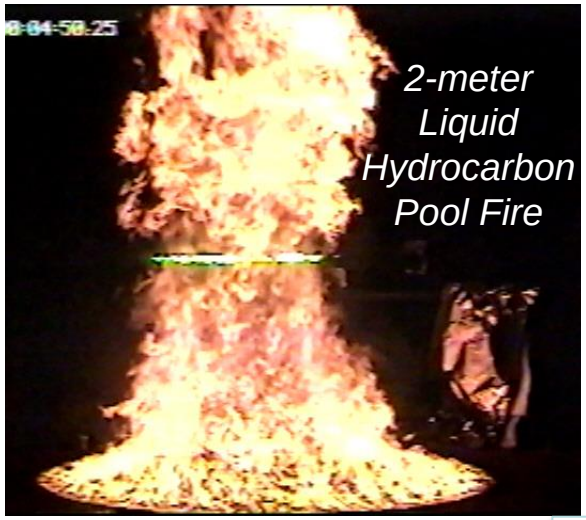
- High temporal resolution – 10 ns or better
- High spatial resolution – 10^{-4} – 10^{-5} cm^3
- Most effective in clean, laboratory flames



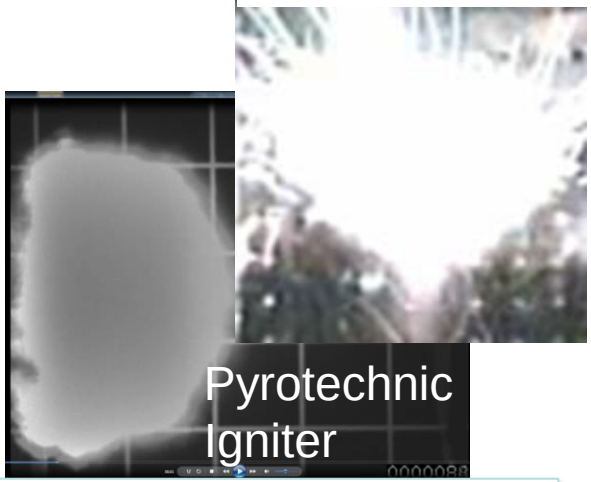


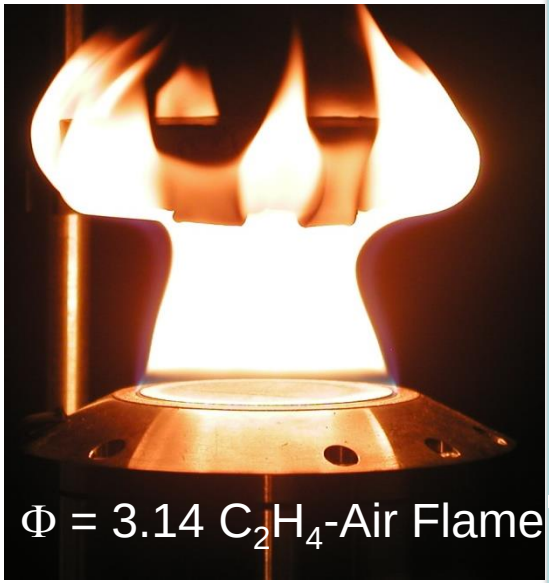
Sandia's application space presents significant challenges

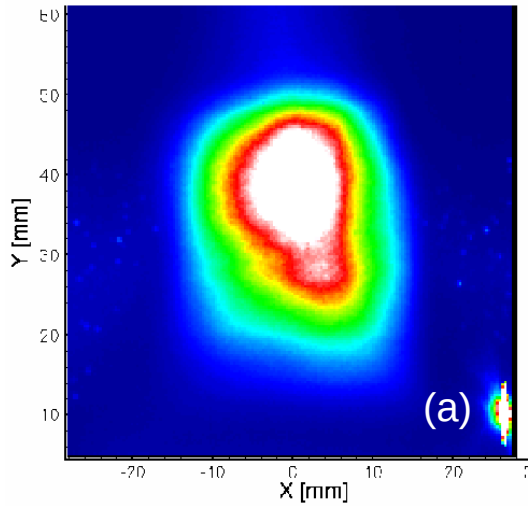
- "Dirty" environments
  - Fire research
  - Energetic materials
- Soot, aluminum particulate
- Luminosity
- Scattering
- Absorption/optical thickness
- ***Large-scale*** of combustion systems



Propellant Burn

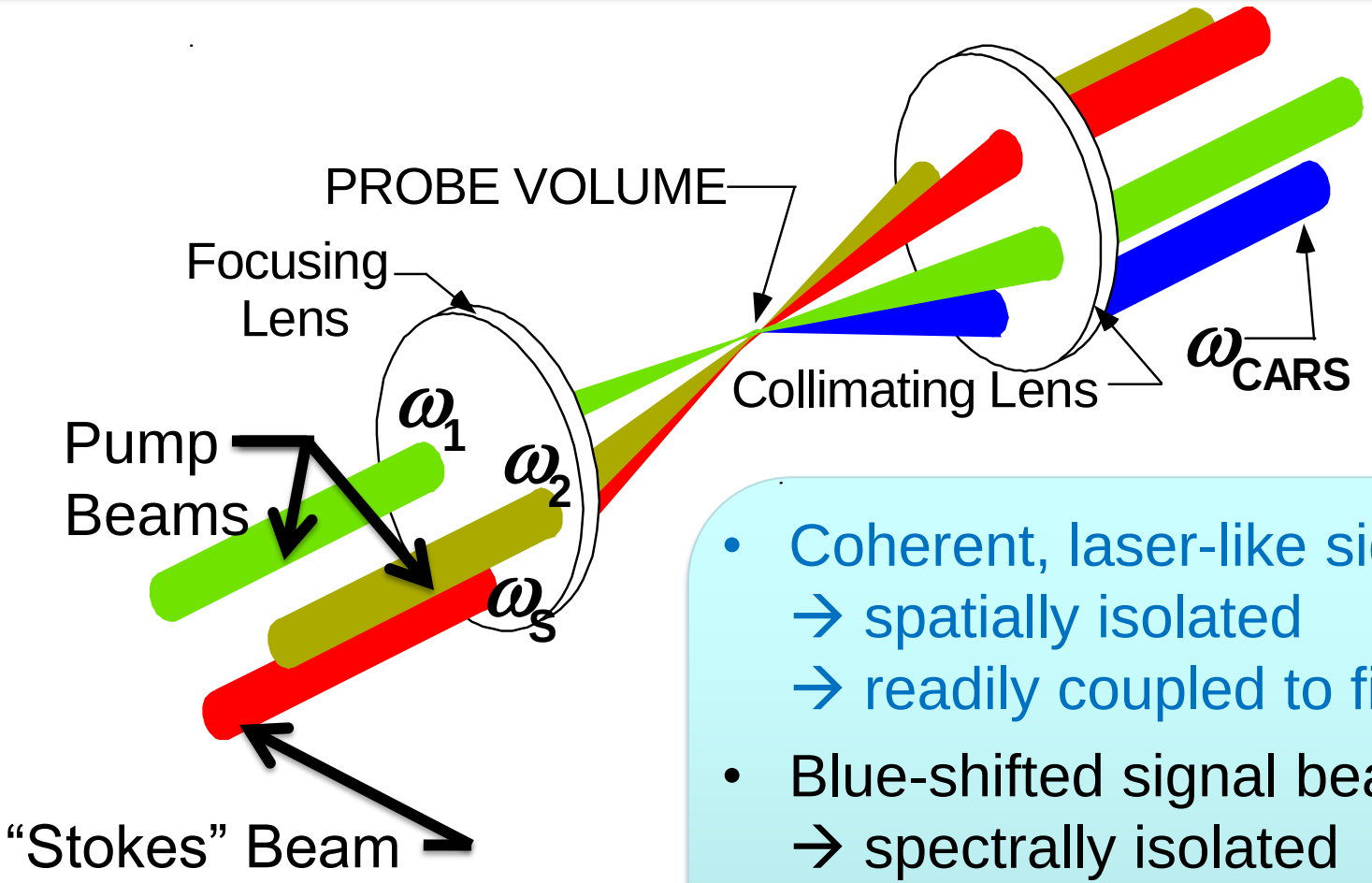






Sooting

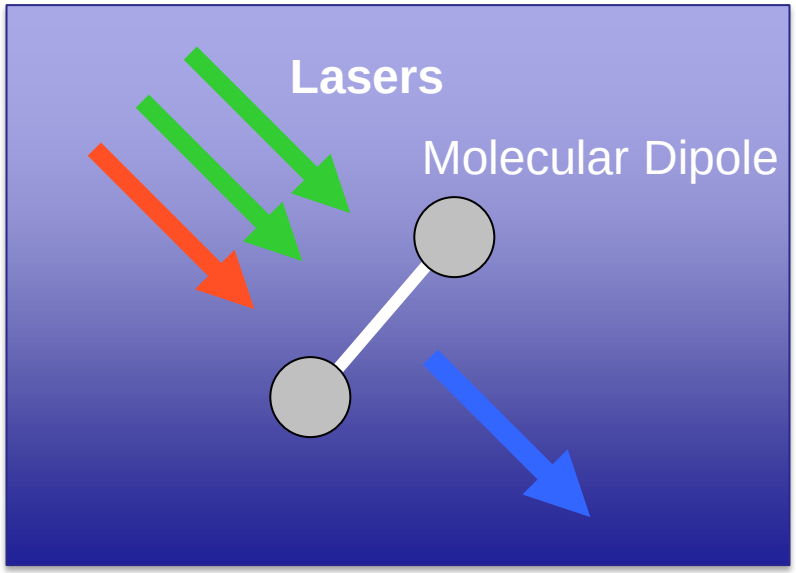
Coherent anti-Stokes Raman Scattering (CARS)



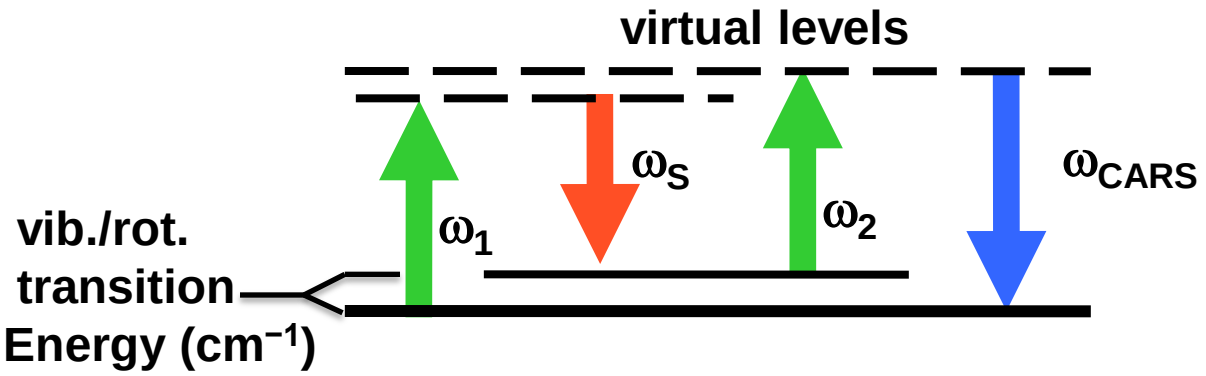
- Coherent, laser-like signal beam
  → spatially isolated
  → readily coupled to fibers
- Blue-shifted signal beam
  → spectrally isolated
- Orders of magnitude stronger than incoherent scattering

CARS Physical Processes: Light/Matter Interaction

- 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

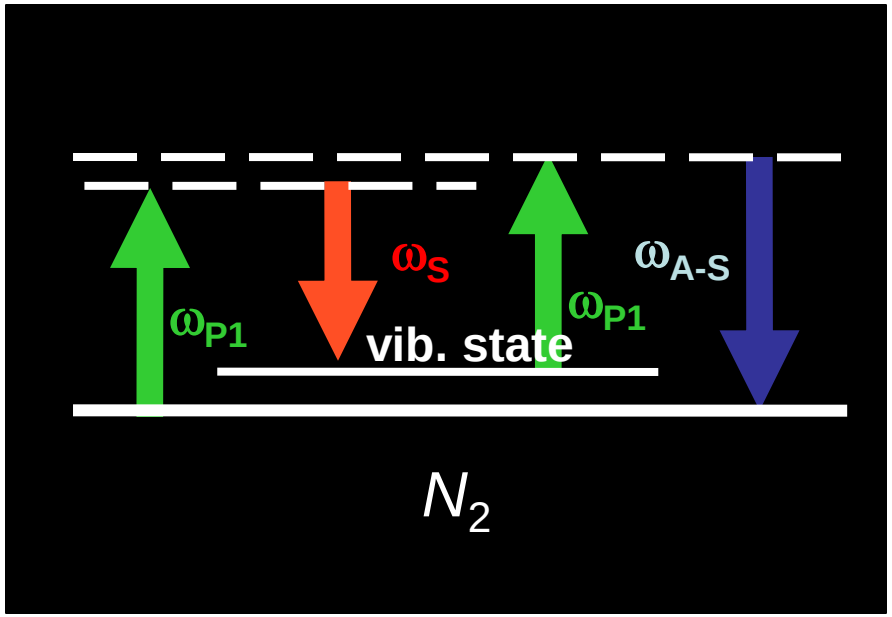


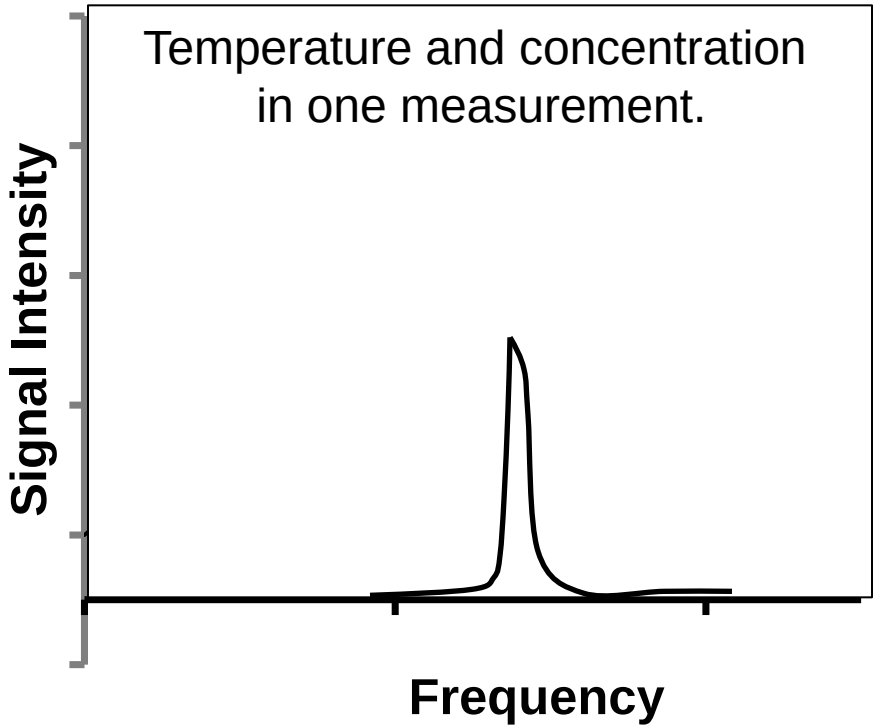
Coherent Anti-Stokes Raman



A broadband source permits single-shot detection

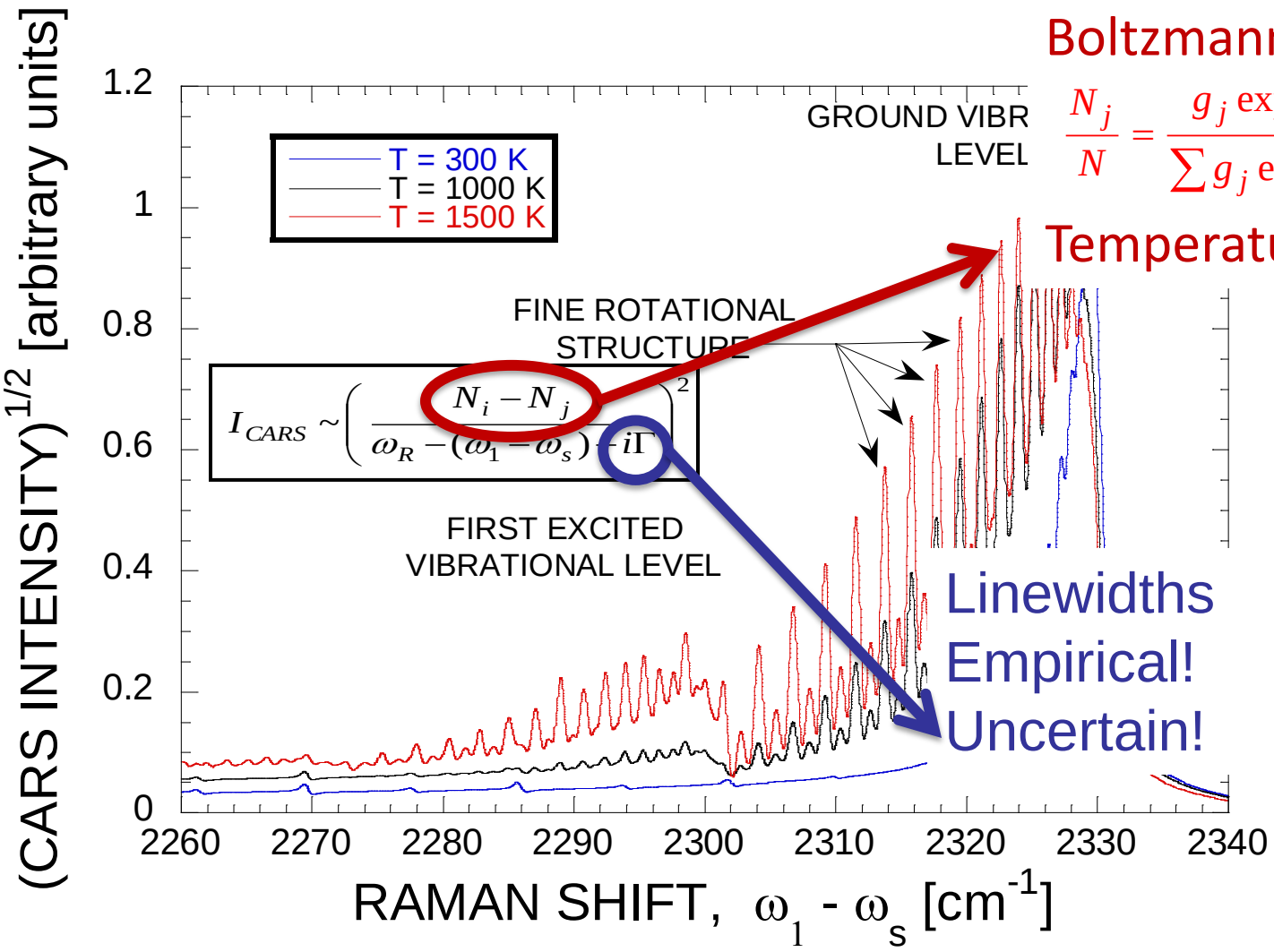
- If all lasers are narrowband one energy level is probed

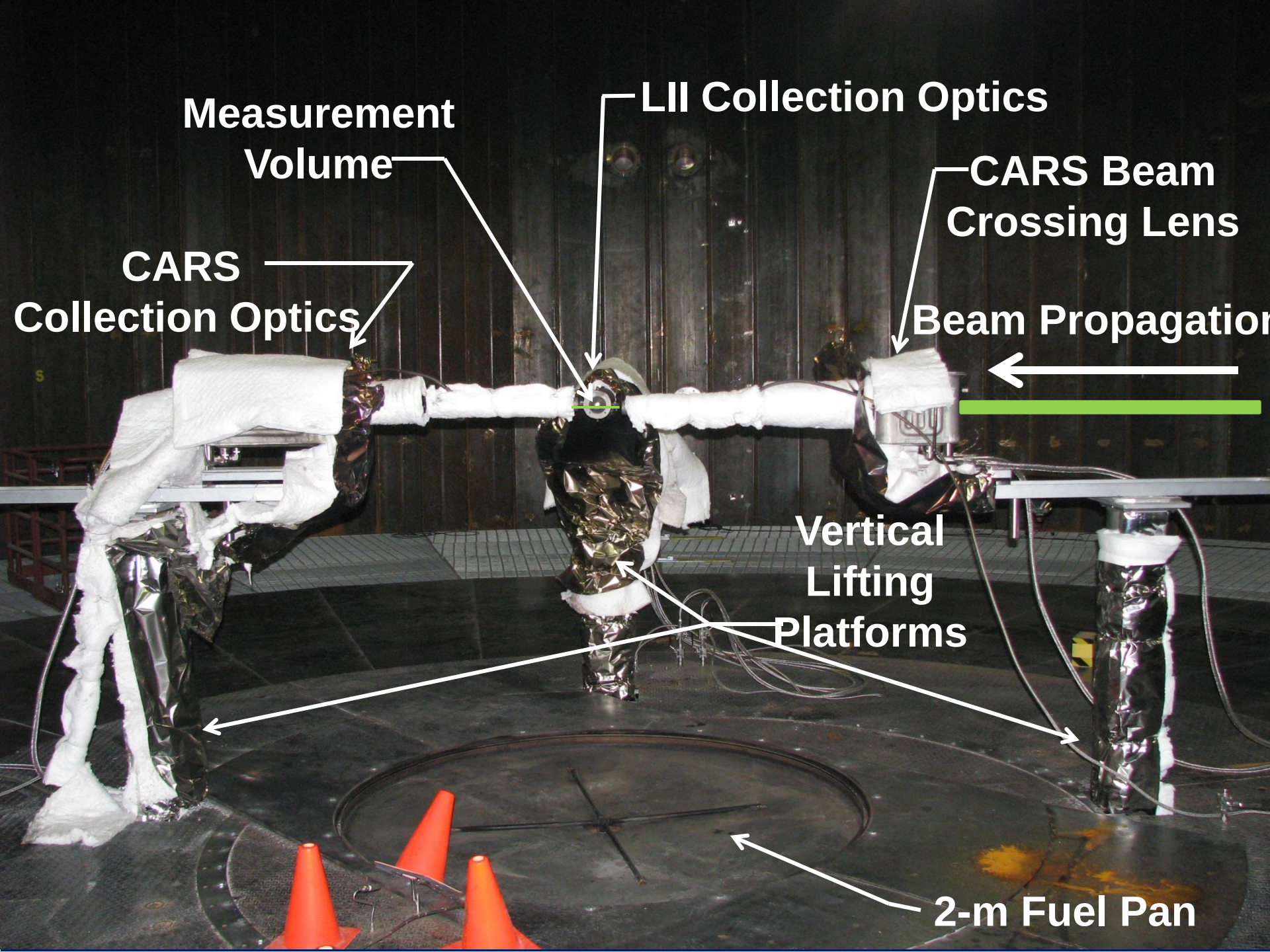




- If one (or more) laser is broadband then a range of energy levels differences are probed

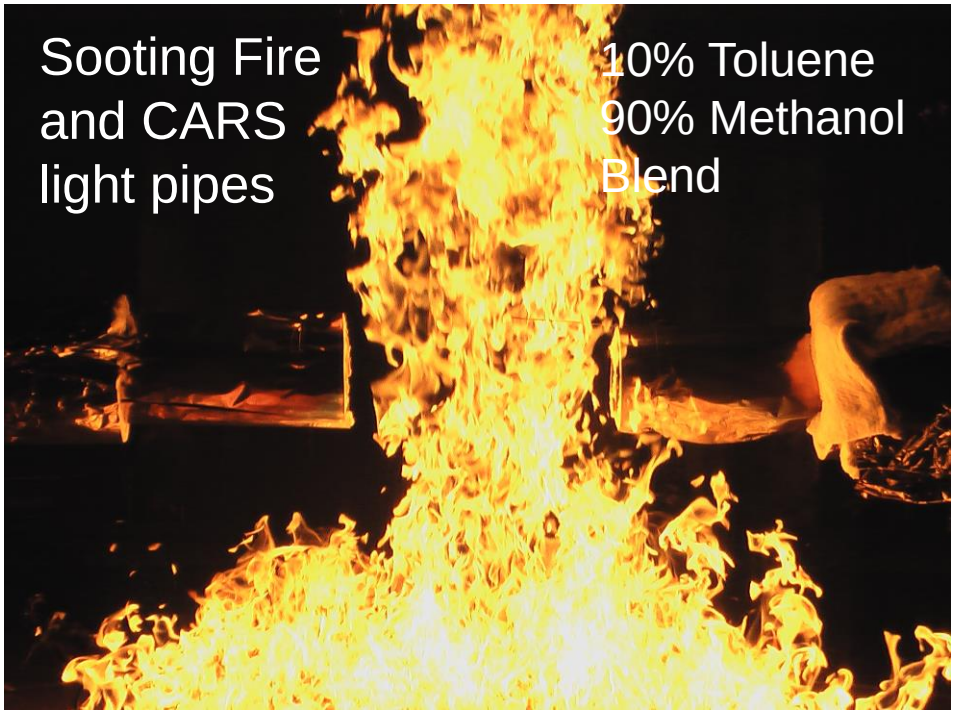
Temperature sensitivity comes from the spectral shape

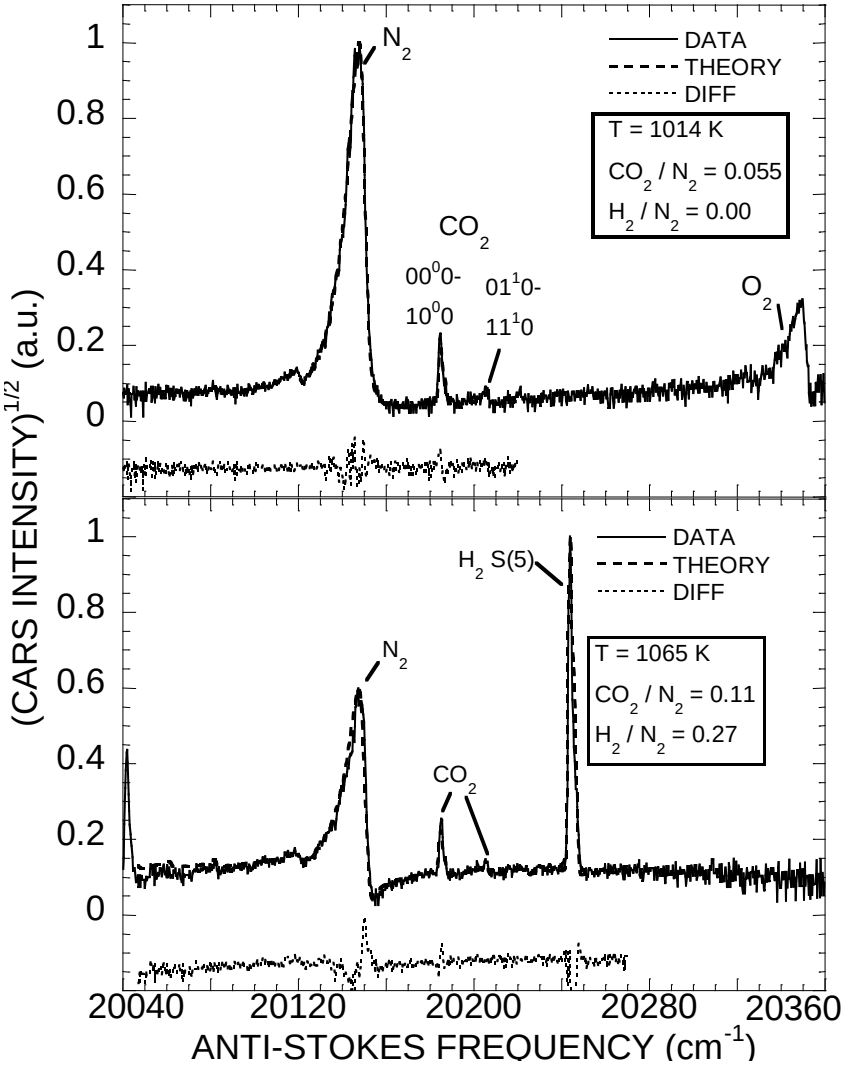



Measurement Volume
LII Collection Optics
CARS Beam Crossing Lens
CARS Collection Optics
Beam Propagation
Vertical Lifting Platforms
2-m Fuel Pan

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

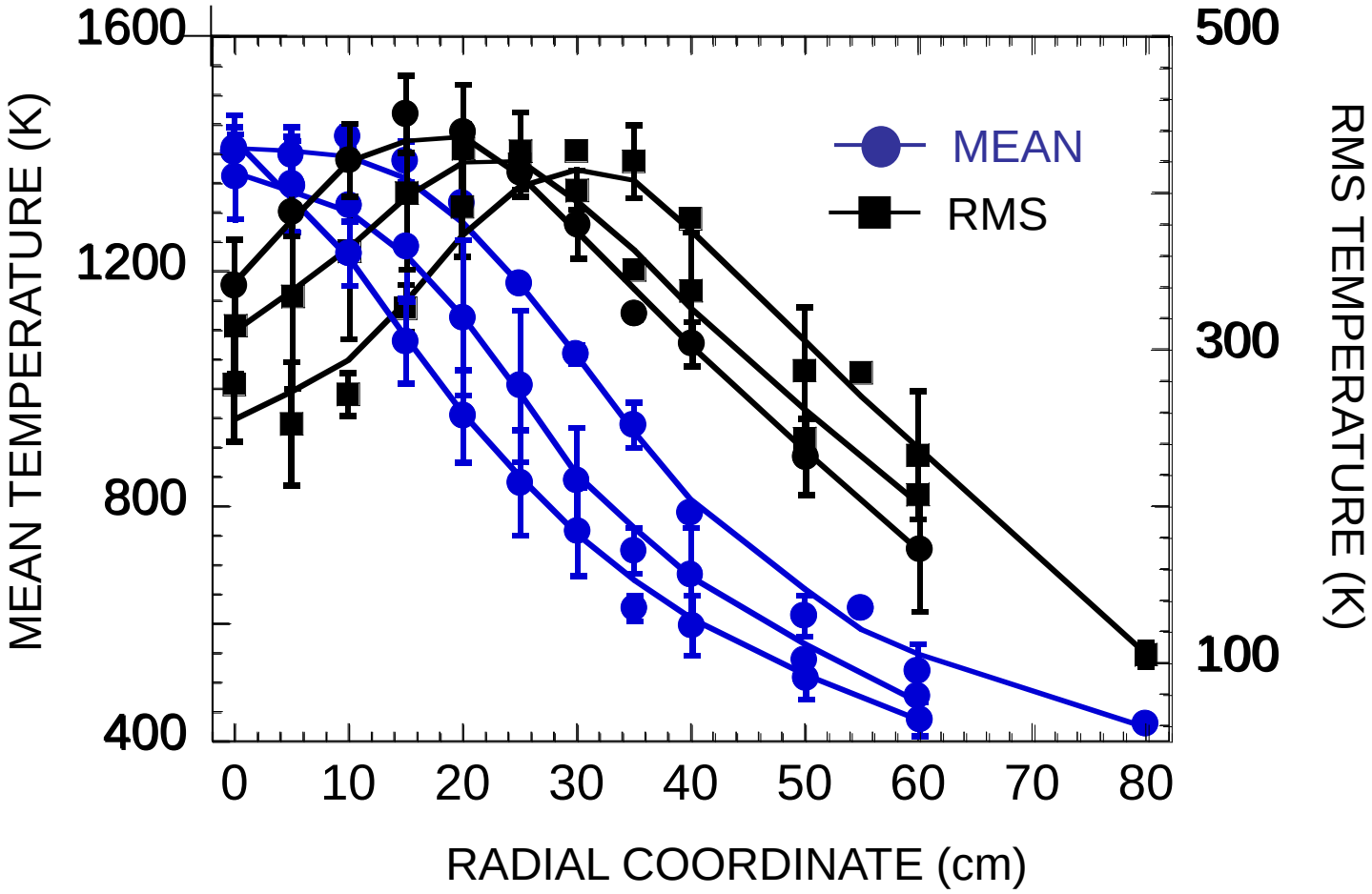
- CARS spectra from sooting fire show N_2,CO_2, H_2, and O_2
- Full ensemble of species data not available as of yet
- Two representative spectra with theoretical fits (Sandia CARSFT code) shown here



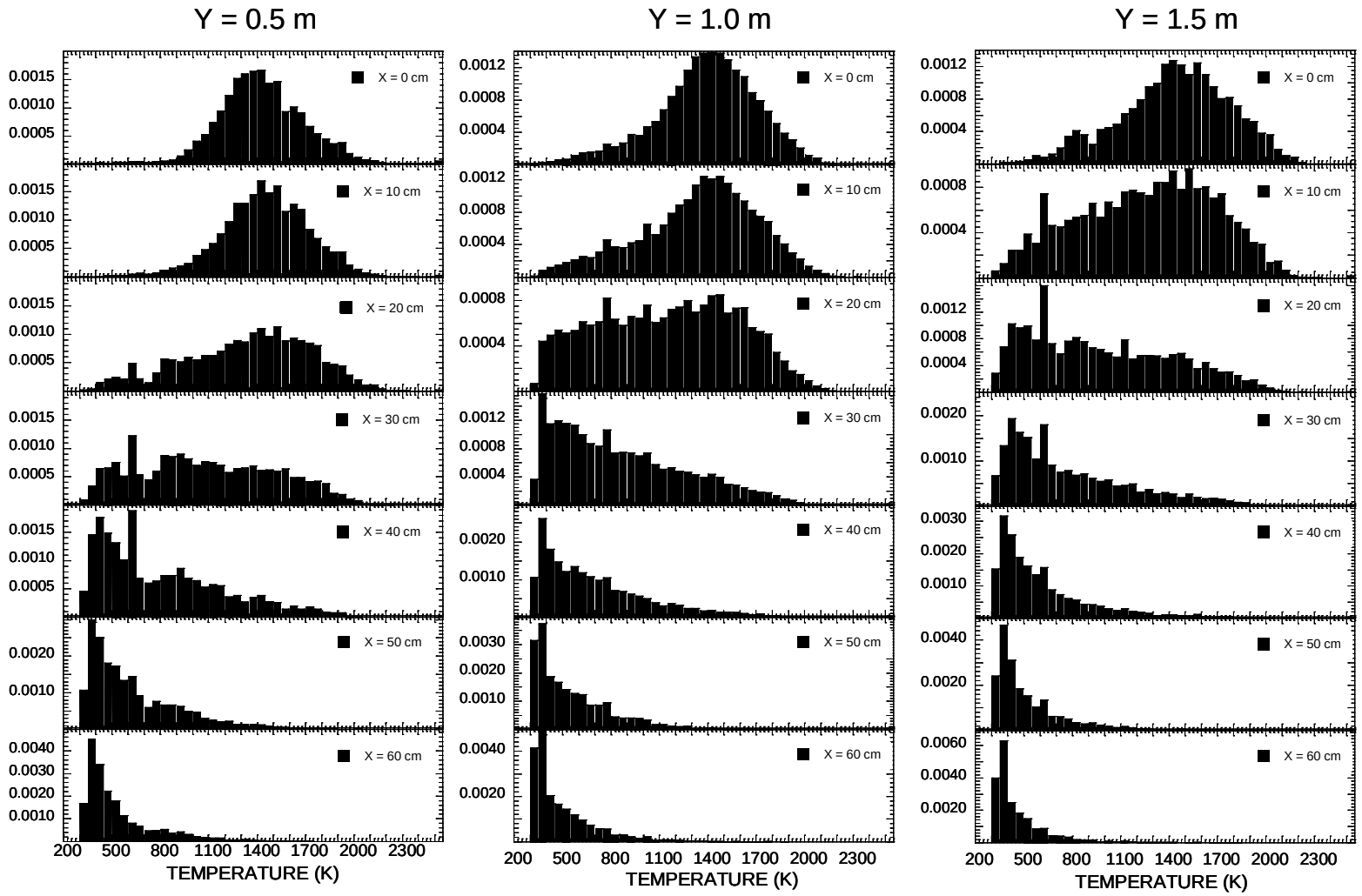


Kearney *et al.*, Proc. Combust. Inst. **32**, 871-878, 2009.

Radial Temperature Profiles – Sooting Pool Fire



Temperature PDF – Sooting Pool Fire



Joint/Temperature Soot Statistics for Emission

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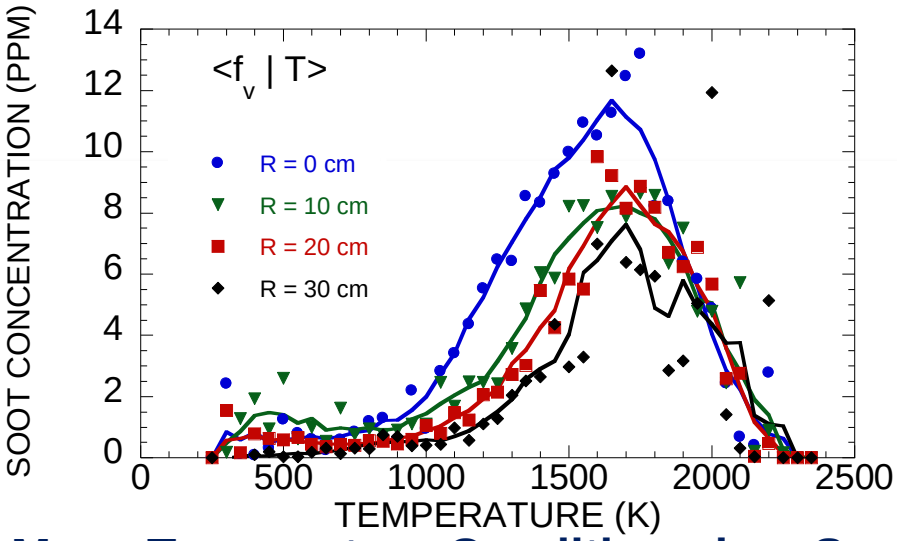
Radiative Transfer Equation

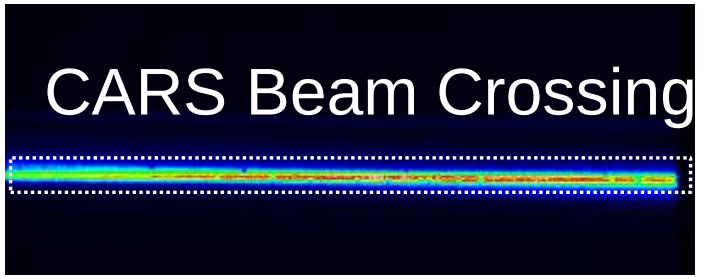
$$\frac{d\overline{I_\lambda}}{ds} = \overline{\mu_\lambda I_{\lambda,b}(T)} - \overline{\mu_\lambda}\,\overline{I_\lambda}$$

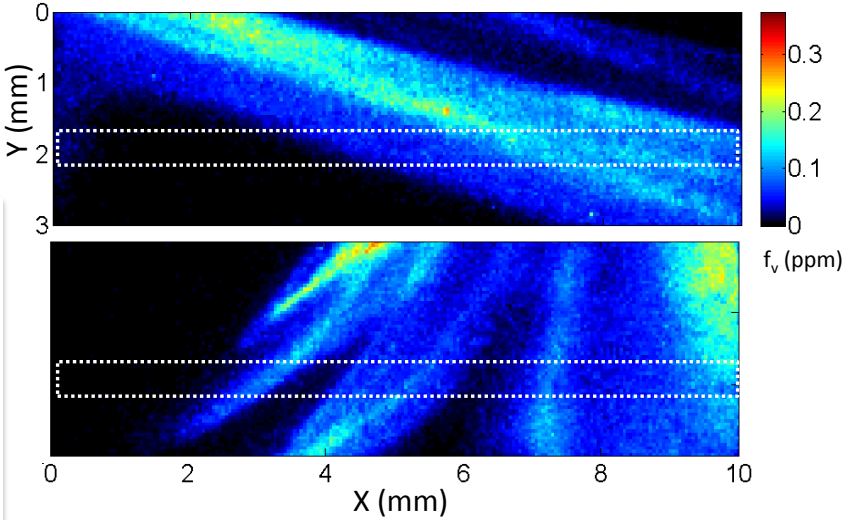
Joint Temp. Soot Statistics Desired

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



Mean Temperature Conditioned on Soot





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

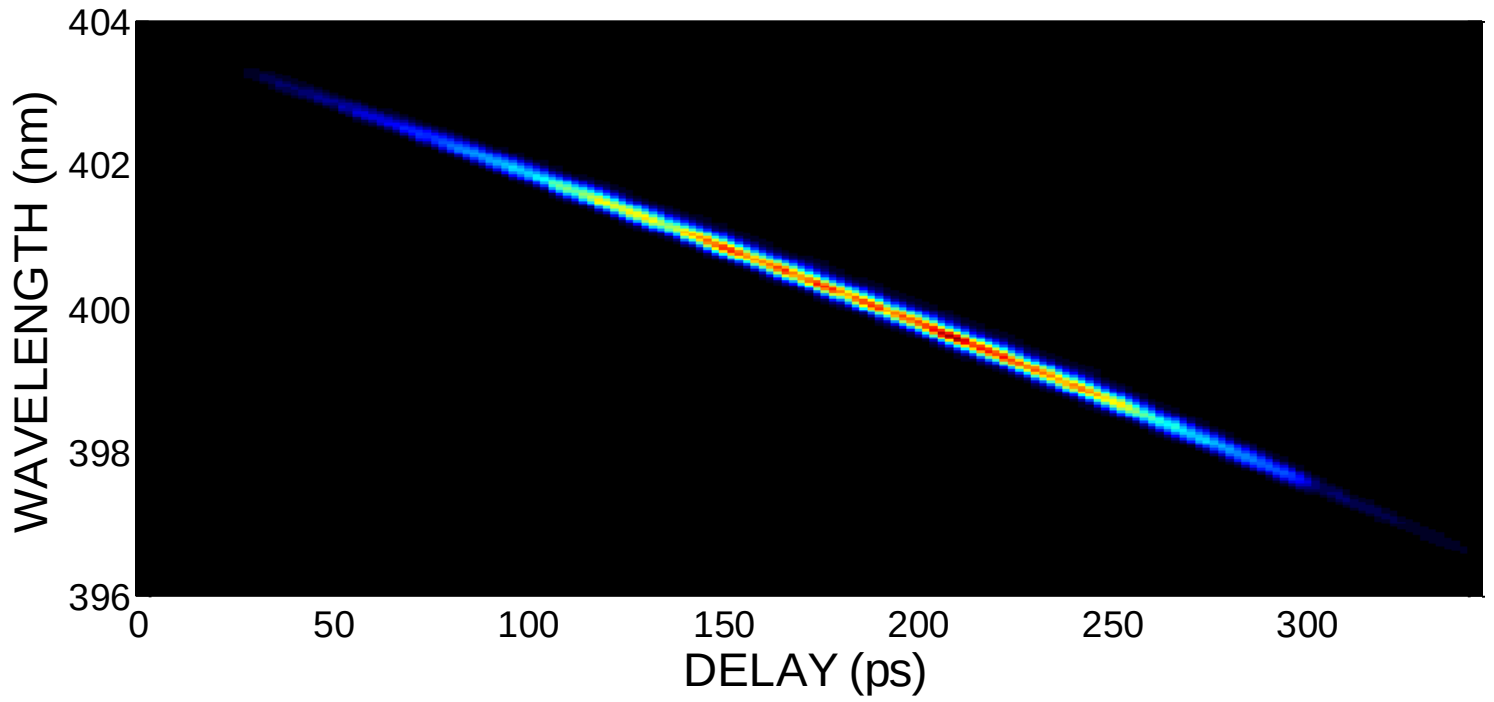
Femtosecond CARS

Why Ultrafast?

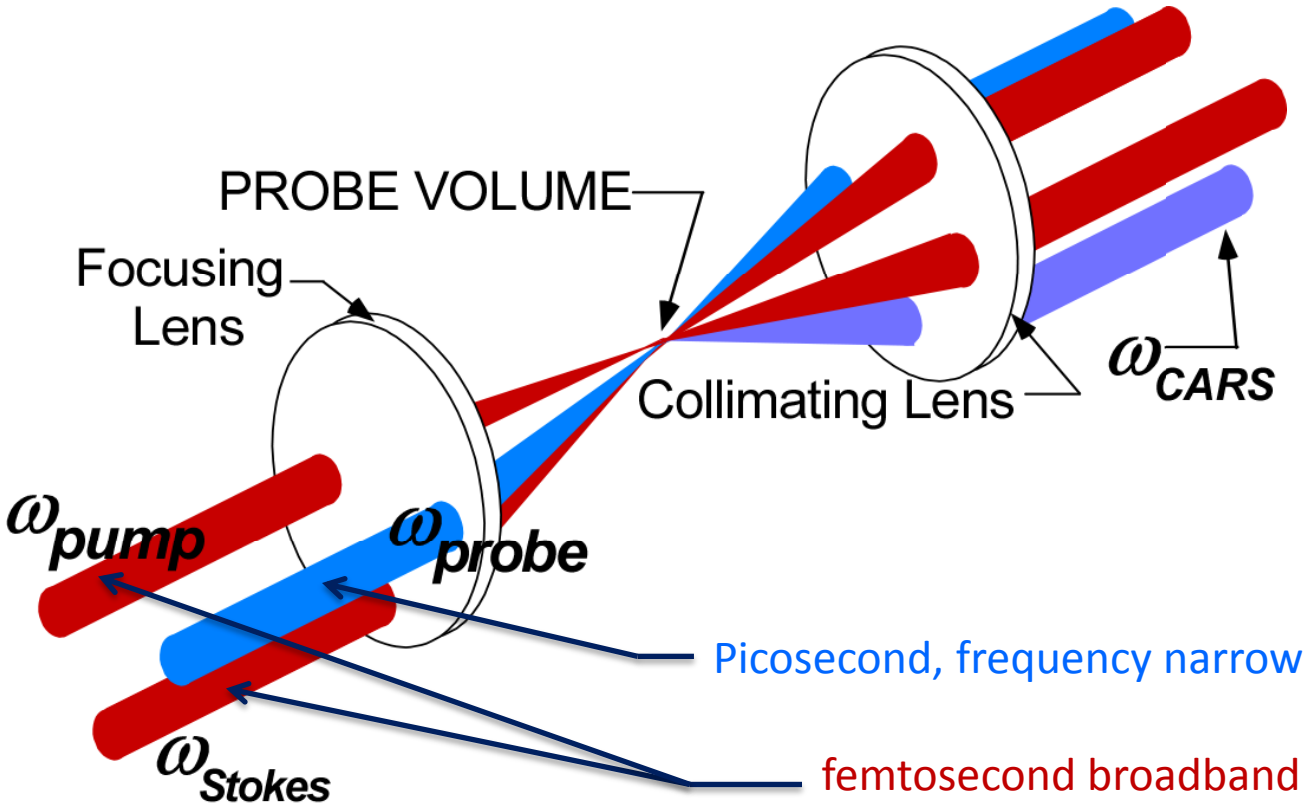
- High-quality (transform-limited) ***broadband*** sources

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

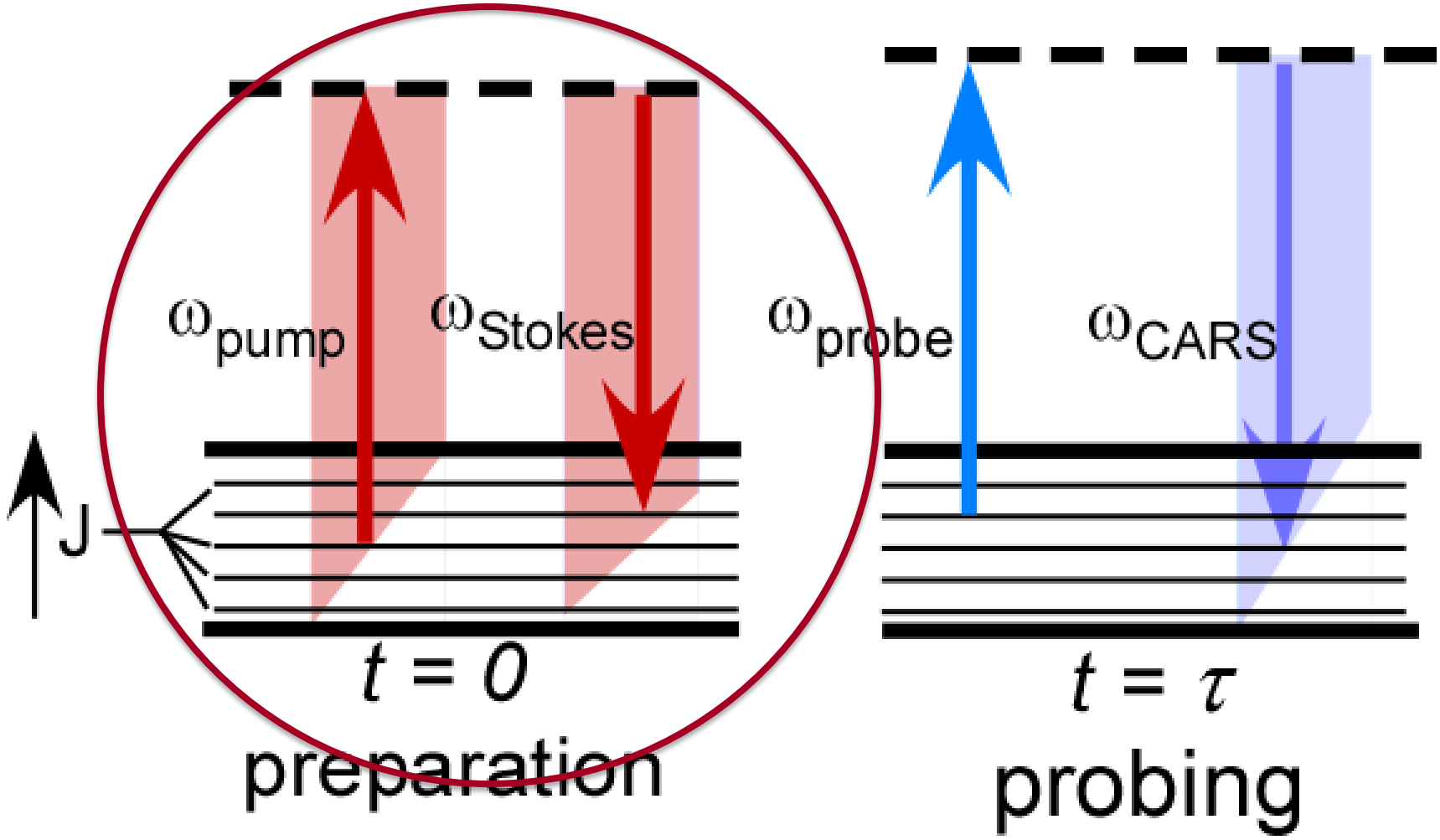
- High repetition rates (kHz vs 10 Hz)
- Transient vs. steady state measurements → no linewidths!



fs/ps ***Rotational*** CARS Experimental Arrangement



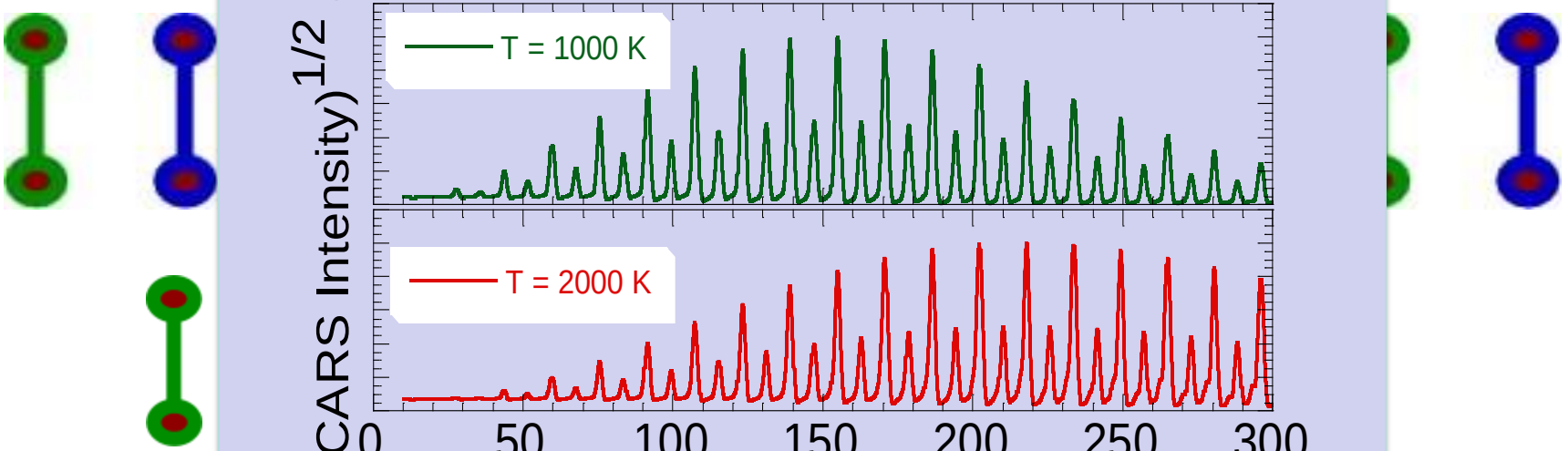
Time-Domain Rotational Raman: $\tau_{laser} << \tau_{molecule}$



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

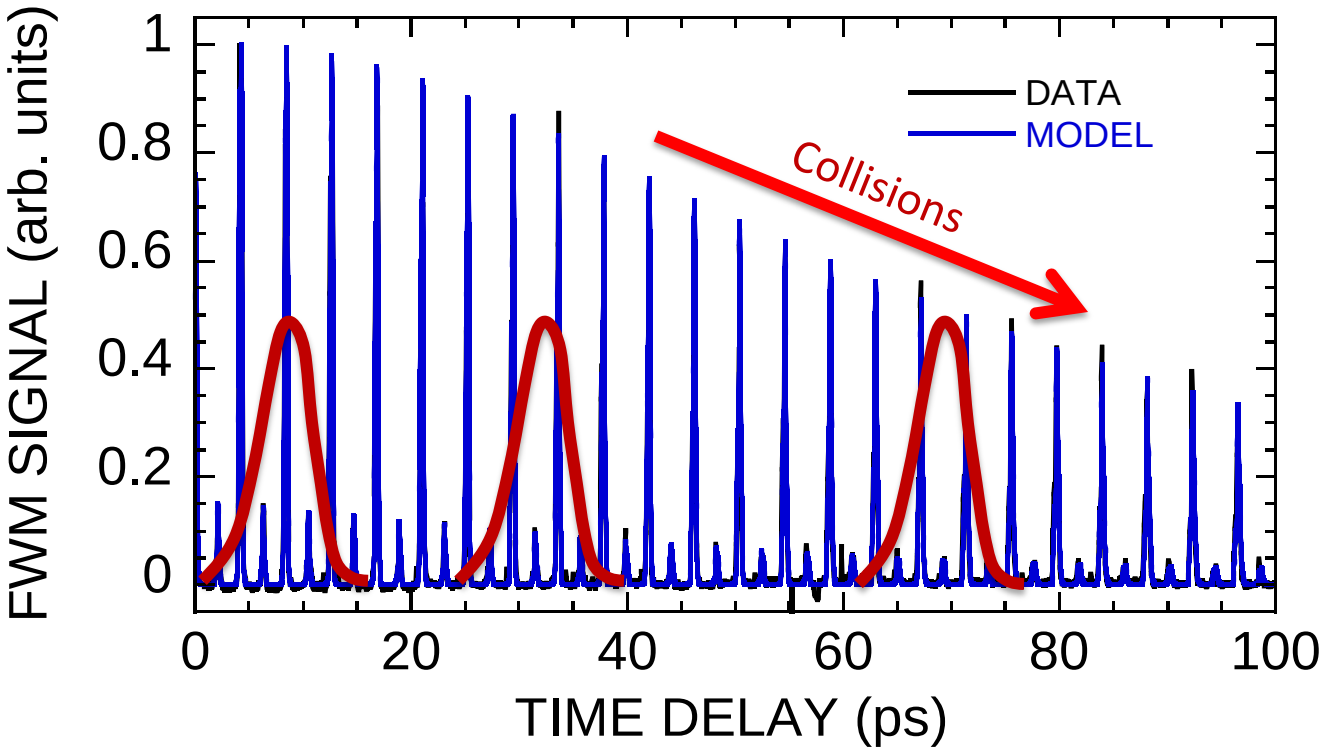
Pump/Stokes Preparation

- Impulsive molecular alignment at time $t = 0$
- Assem … er J states
- Rotor e…
- Rotors …



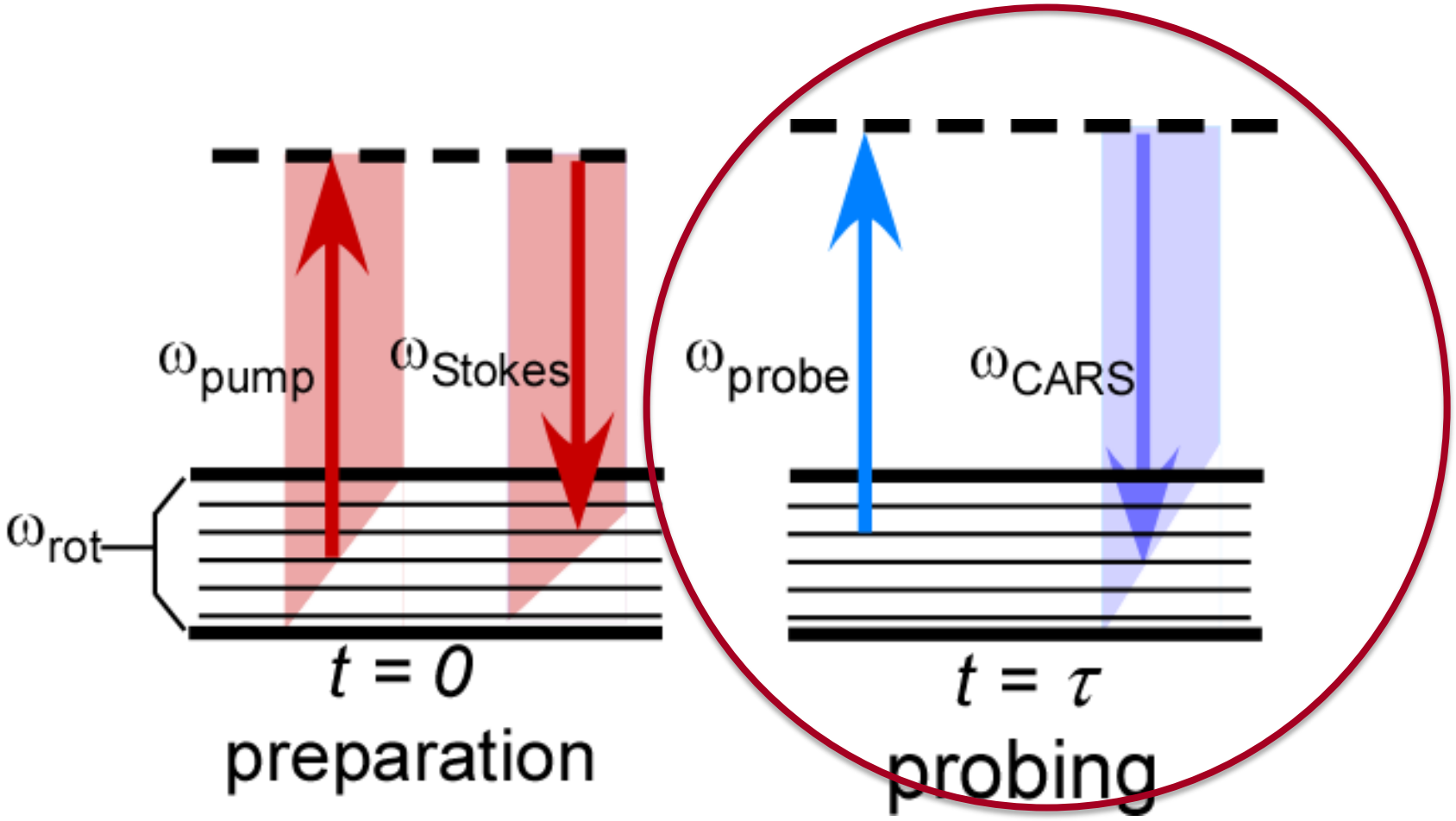
- Assen… …on
- The result is a periodically recurring and long-lived Raman polarization

Measured Response in N_2 at T = 300 K



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

Probe Step



Probe Step and Spectral Synthesis

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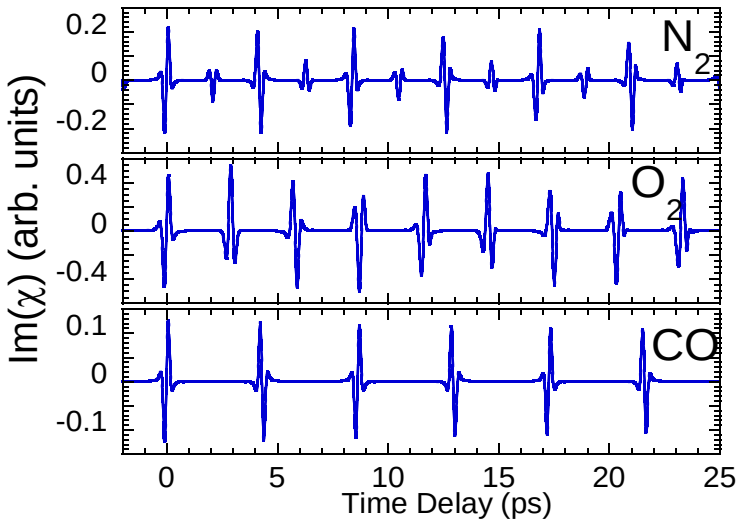
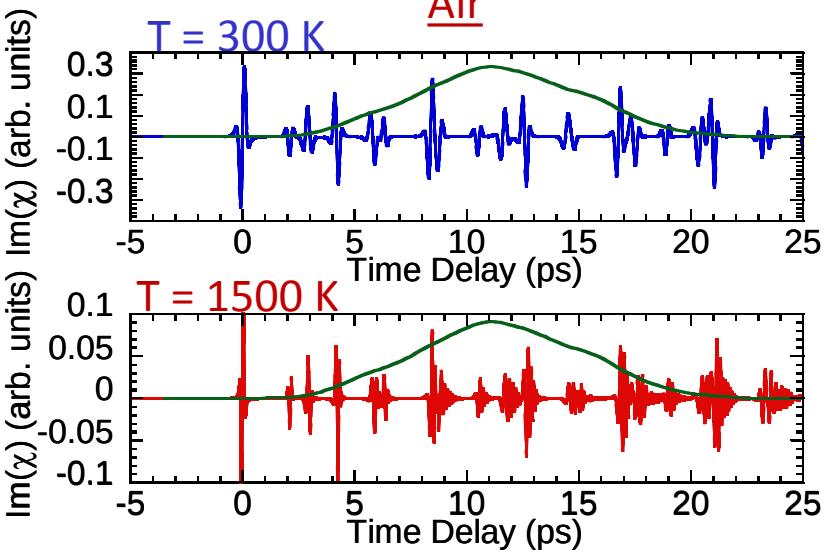
Pure Gases, T = 300 K

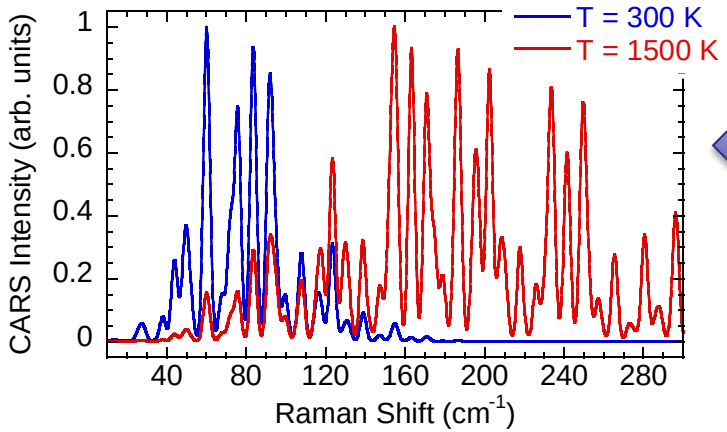


Air



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

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



FFT

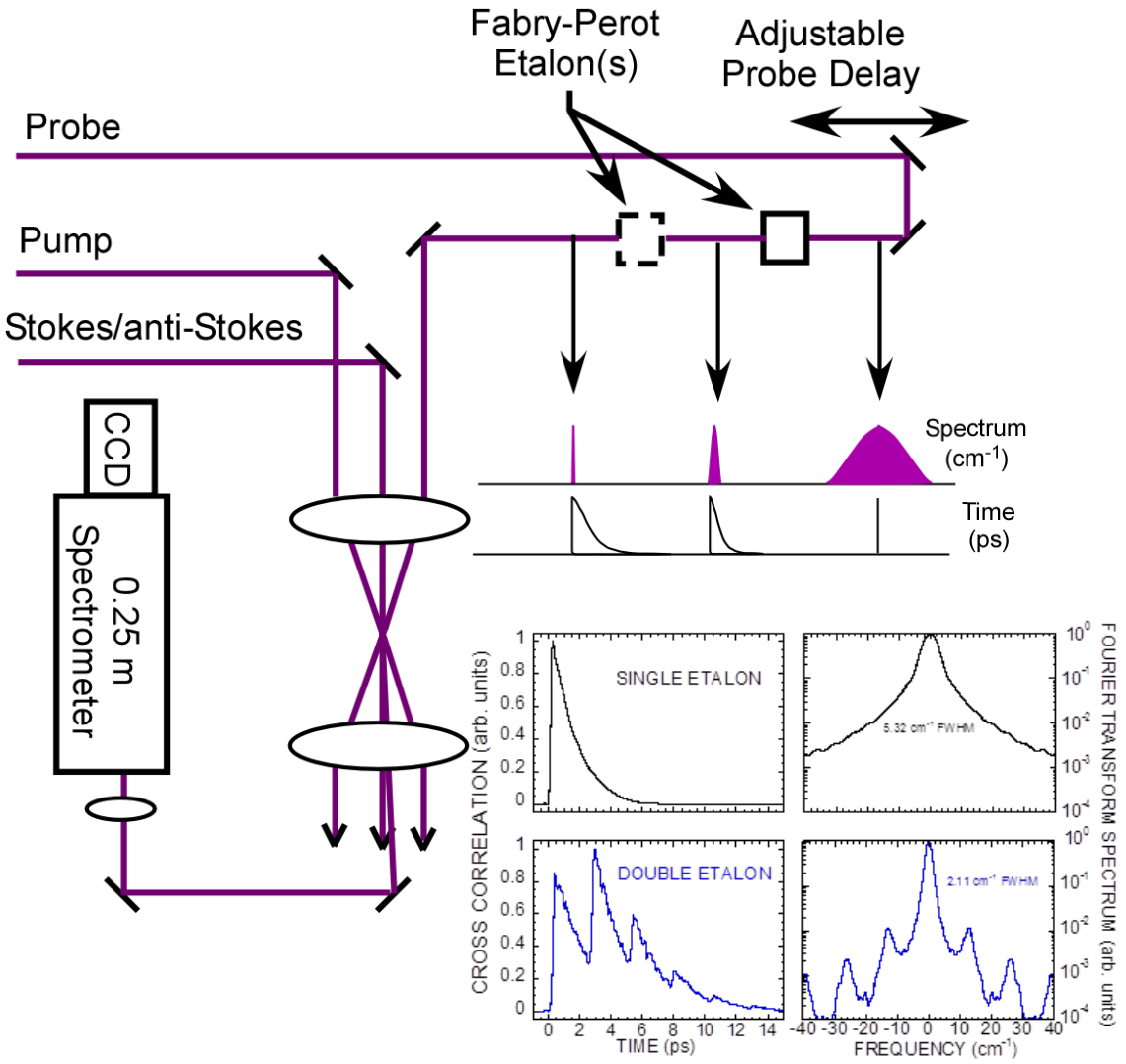
$$E_{CARS}(t) = E_{pr}(t-\tau)\exp\left[i\,\omega_o(t-\tau)\right] \times \chi(t)$$

Proof-of-Concept Experiments in Air

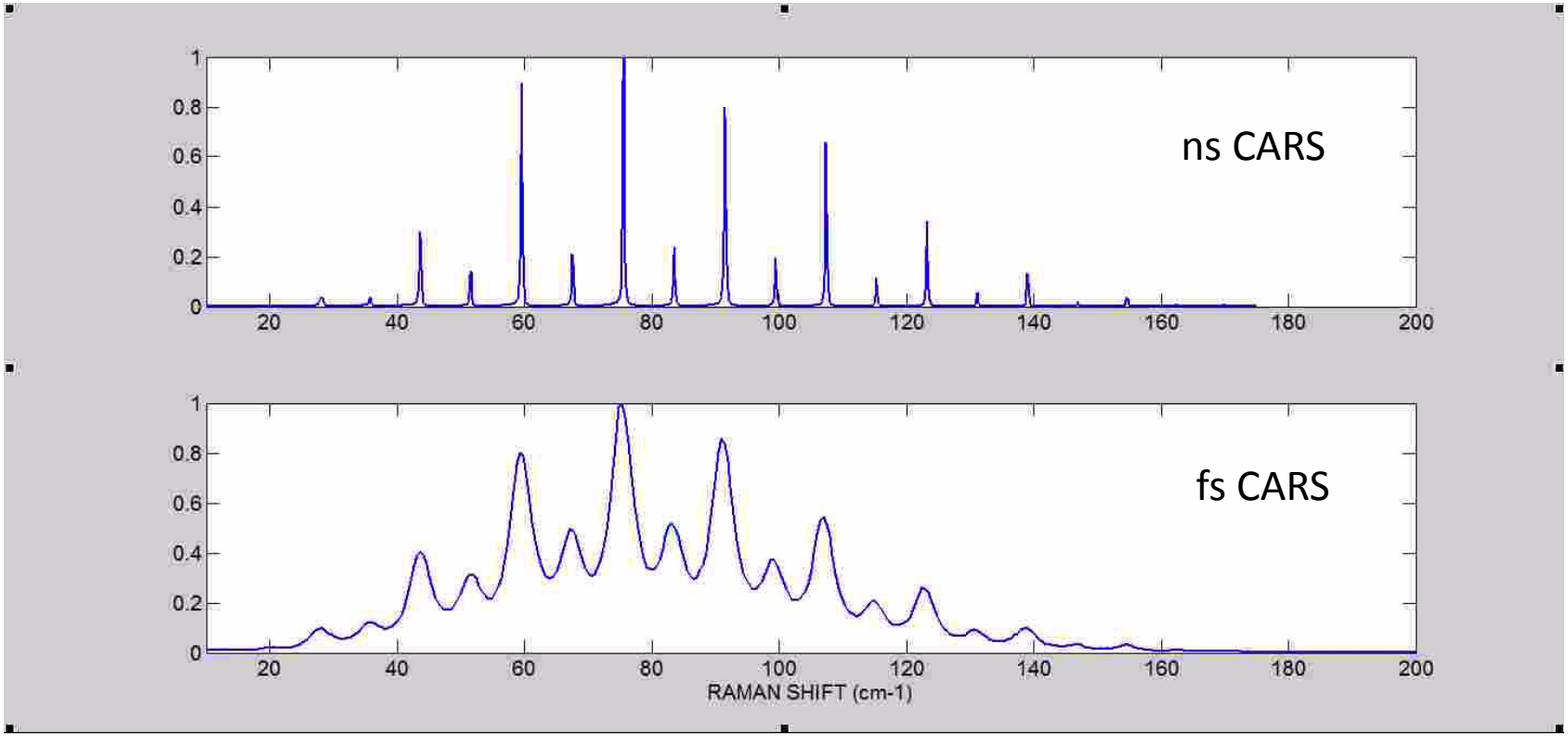
- "Bandwidth-Carving" to generate ps probe pulse
- Raman lines "resolved"

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

- Two different probe resolutions investigated with single- and double-etalon configurations
- Very inefficient (0.8 to 2.4% or less transmission)
- Atmospheric air spectra in tube-furnace up to 800 K for probe delays up to 20 ps

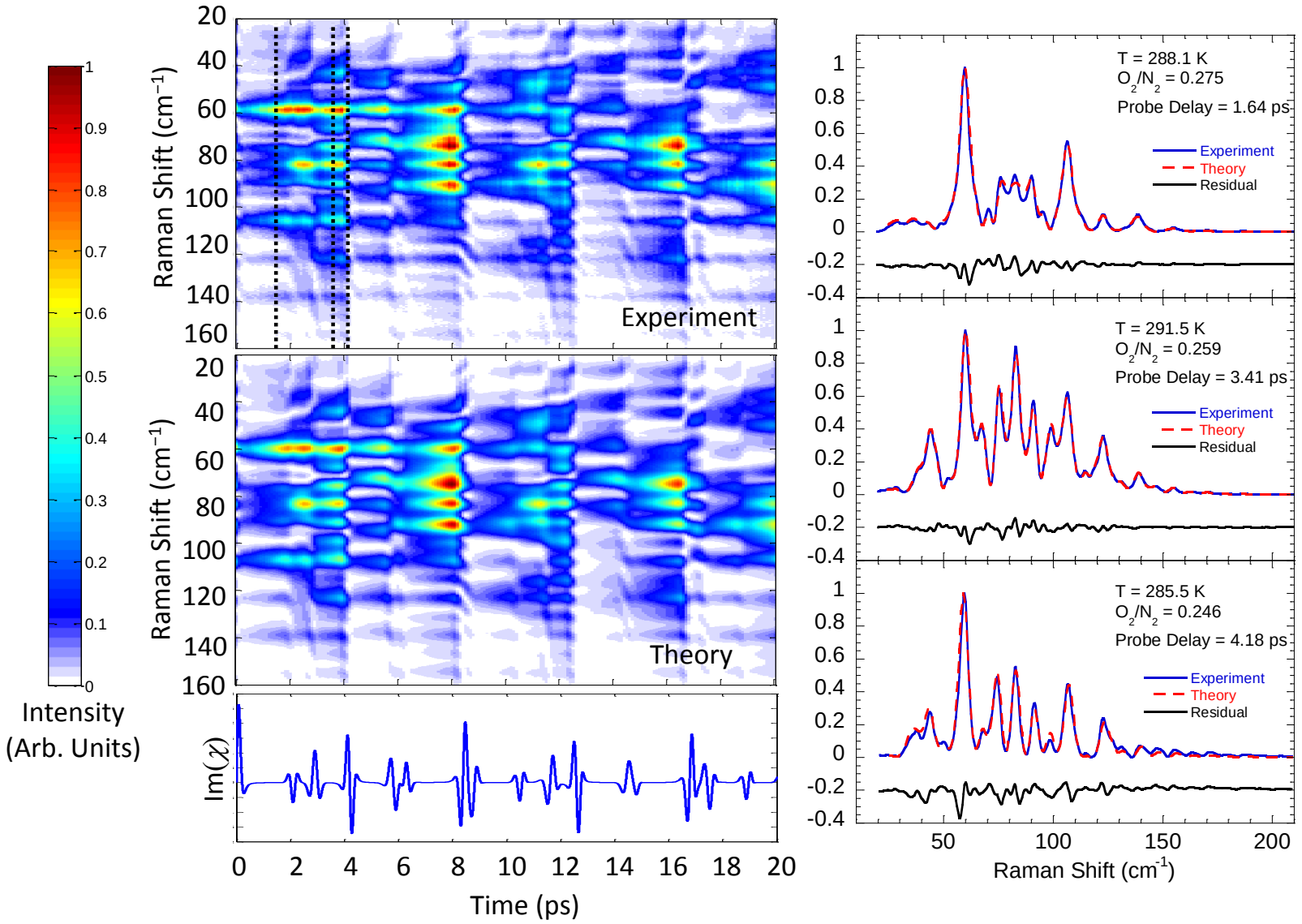


Low noise fs preparation pulses result can result in higher single-laser-shot precision

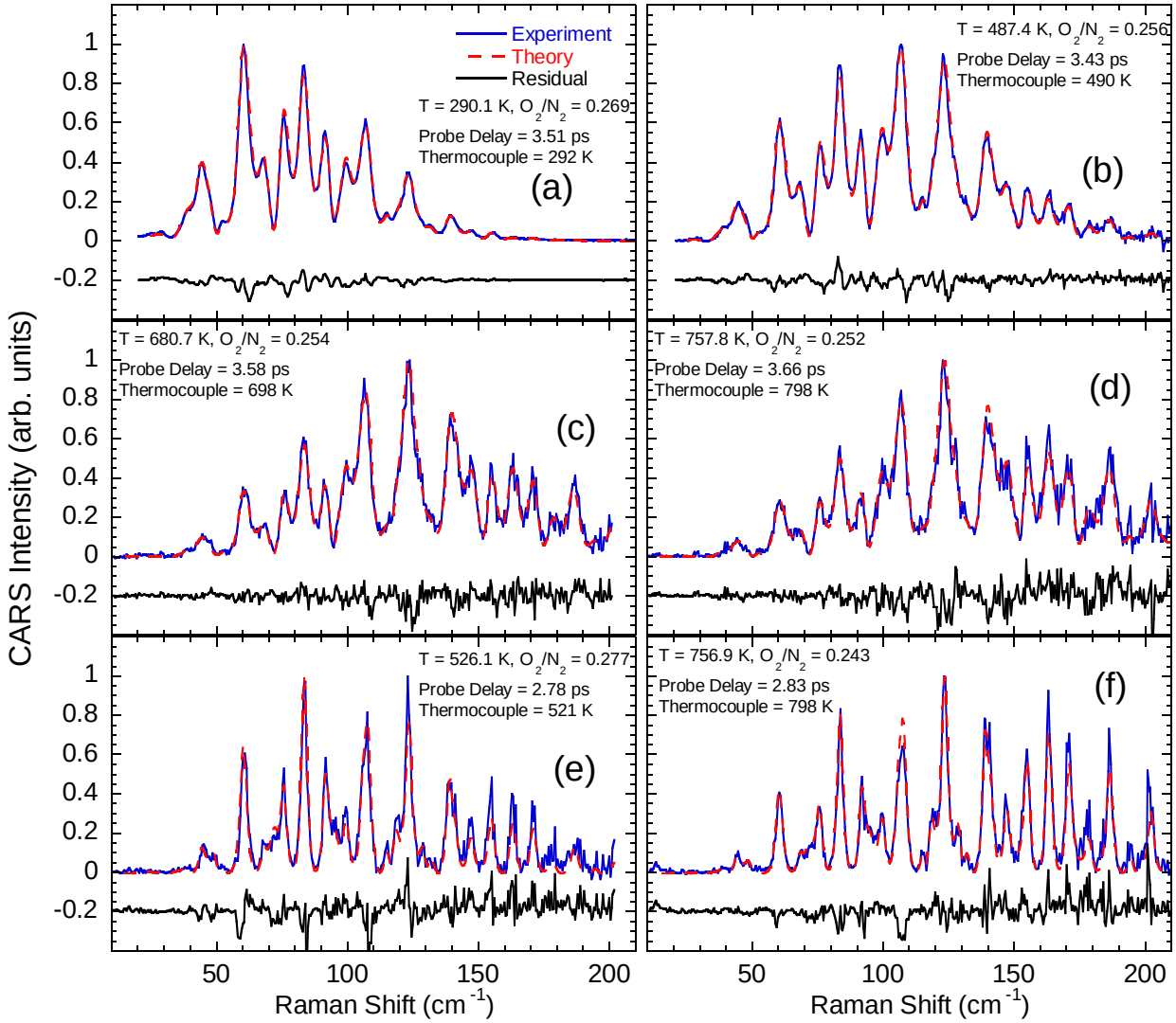


Room-temperature N_2 spectra

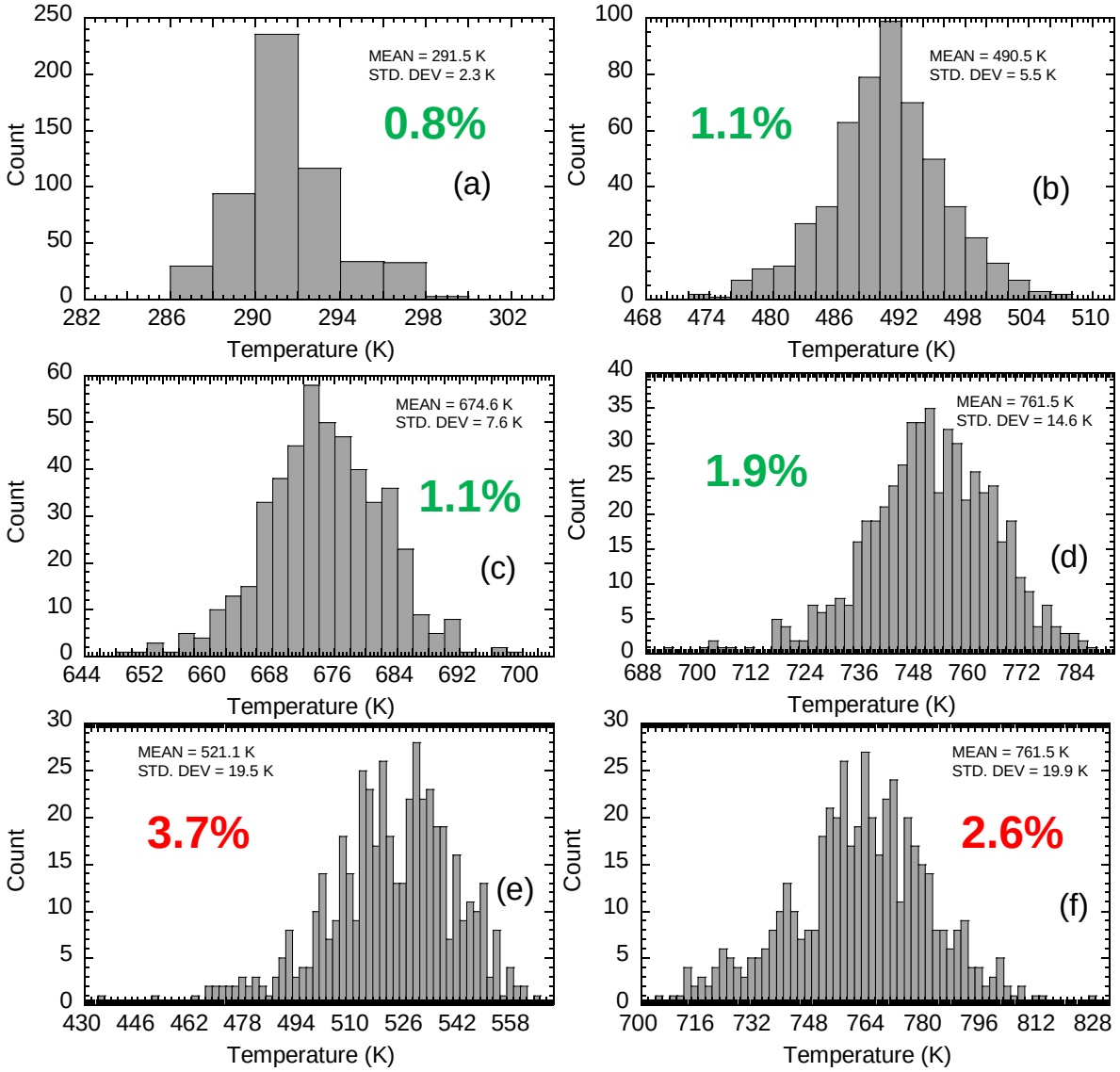
Model validation in room-temperature air: 1.5-ps probe



Collision-free spectra acquired at 1 kHz rate!

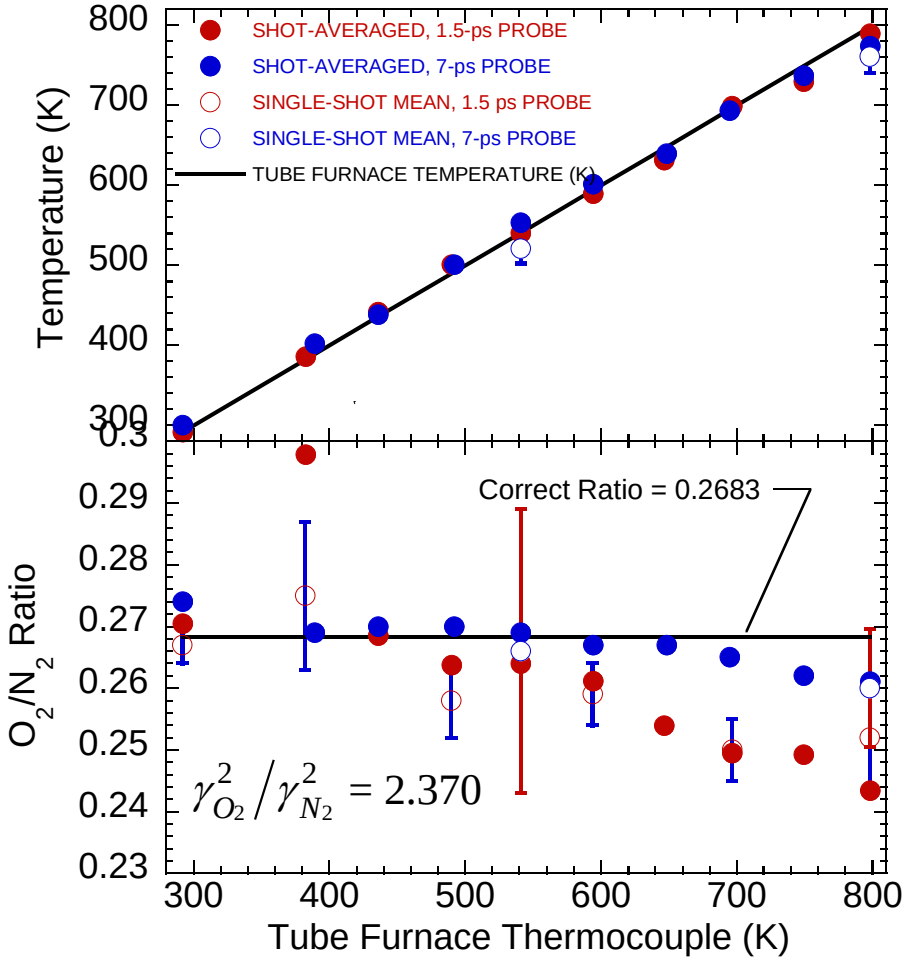


Single-shot temperature histograms



Proof-of-concept: 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



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

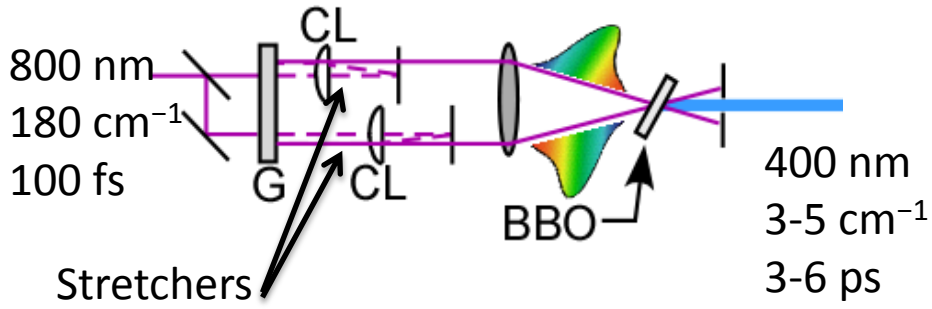
Second-Harmonic Bandwidth Compression (SHBC)

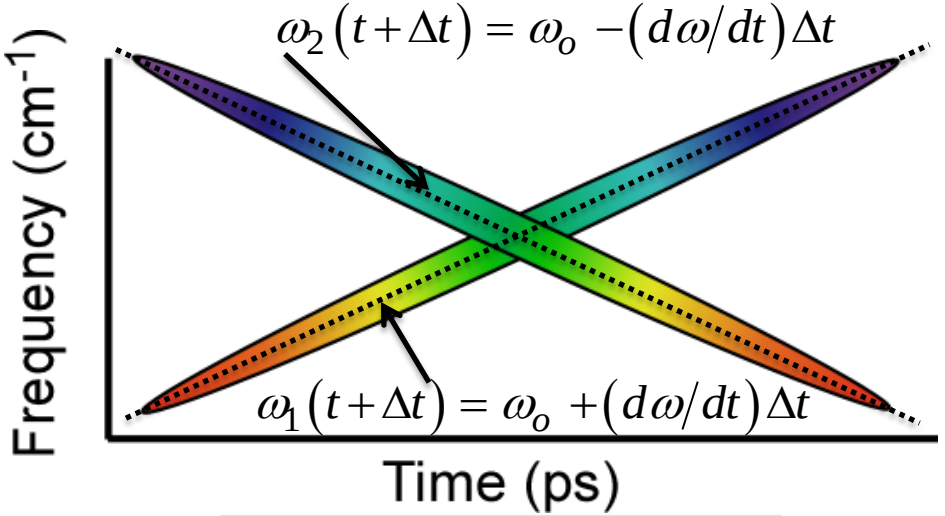
- Commercial device (Light Conversion)
- Converts fs radiation at 800 nm to ps radiation at 400 nm
- Grating pulse stretchers
- **Phase-conjugate** temporal chirps imparted upon broadband fs pumps
- Sum-frequency generation in BBO
- Output linewidth 3.5-4.0 cm^{-1}

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

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

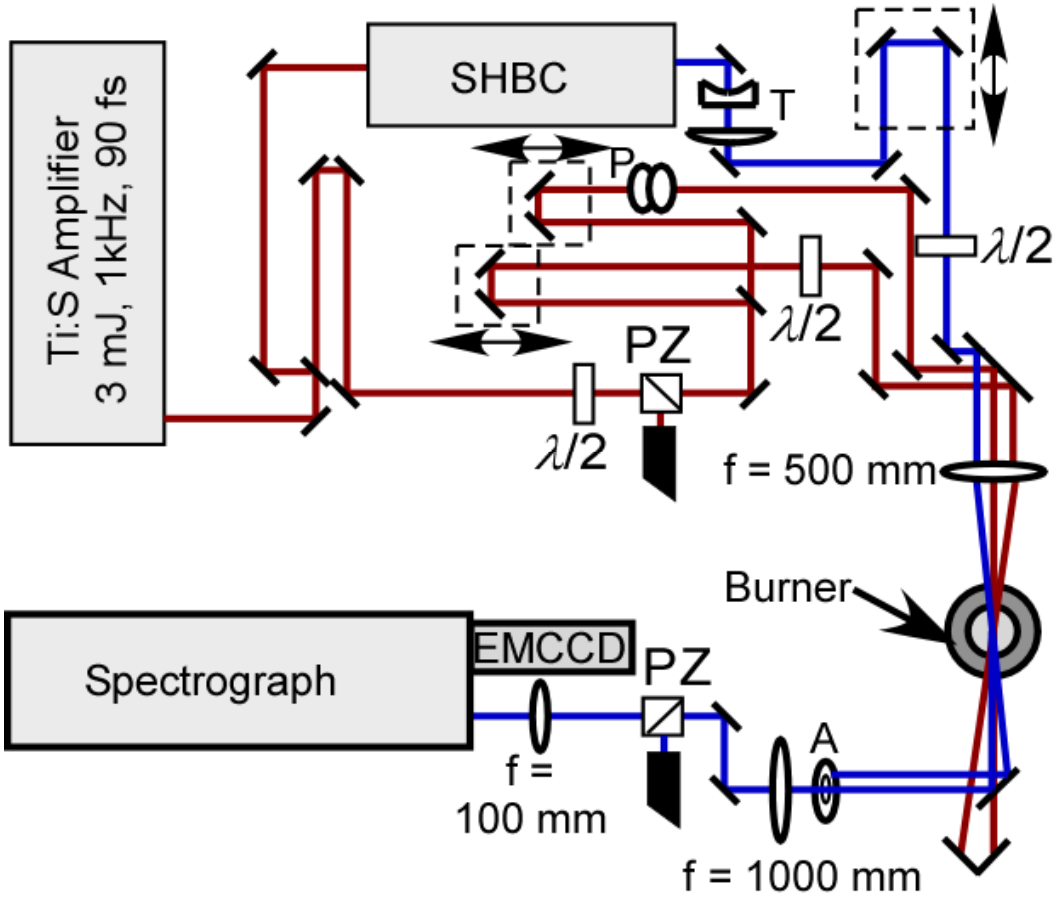
- Conversion efficiency: 35-50%!
- Output pulse energy: 1-1.4 mJ!





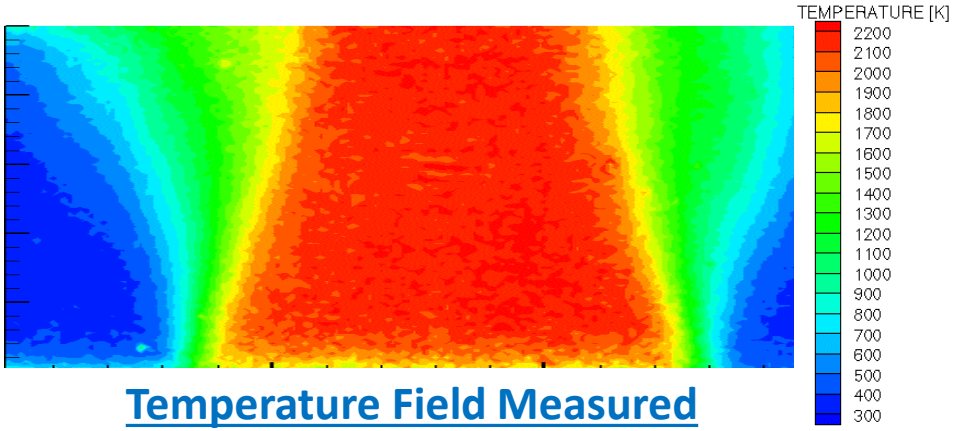
$$\omega_{sfg} = \omega_1 + \omega_2 = 2\omega_o$$

fs/ps CARS Instrument

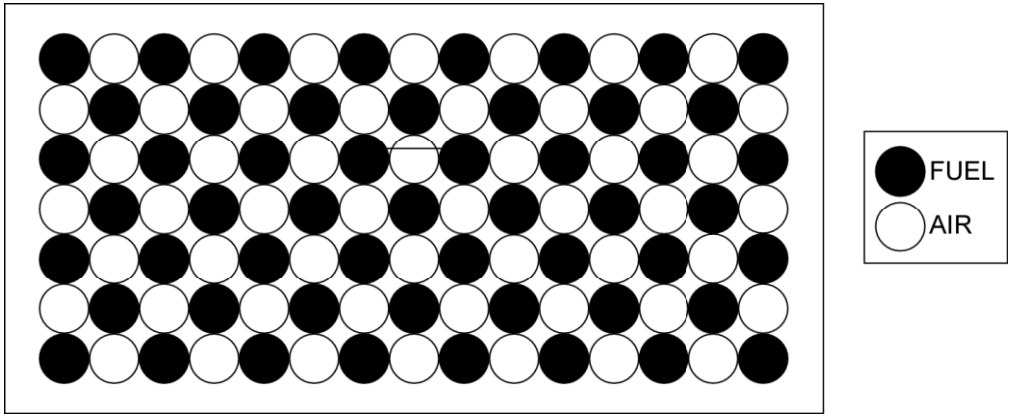


Hencken Burner

- Slightly lifted flat flame
- Flow rates 98 to 116 SLPM
- Non-premixed
- Provides ***nearly adiabatic flames***
- Temperature and major species mole fractions ***calculated from equilibrium***

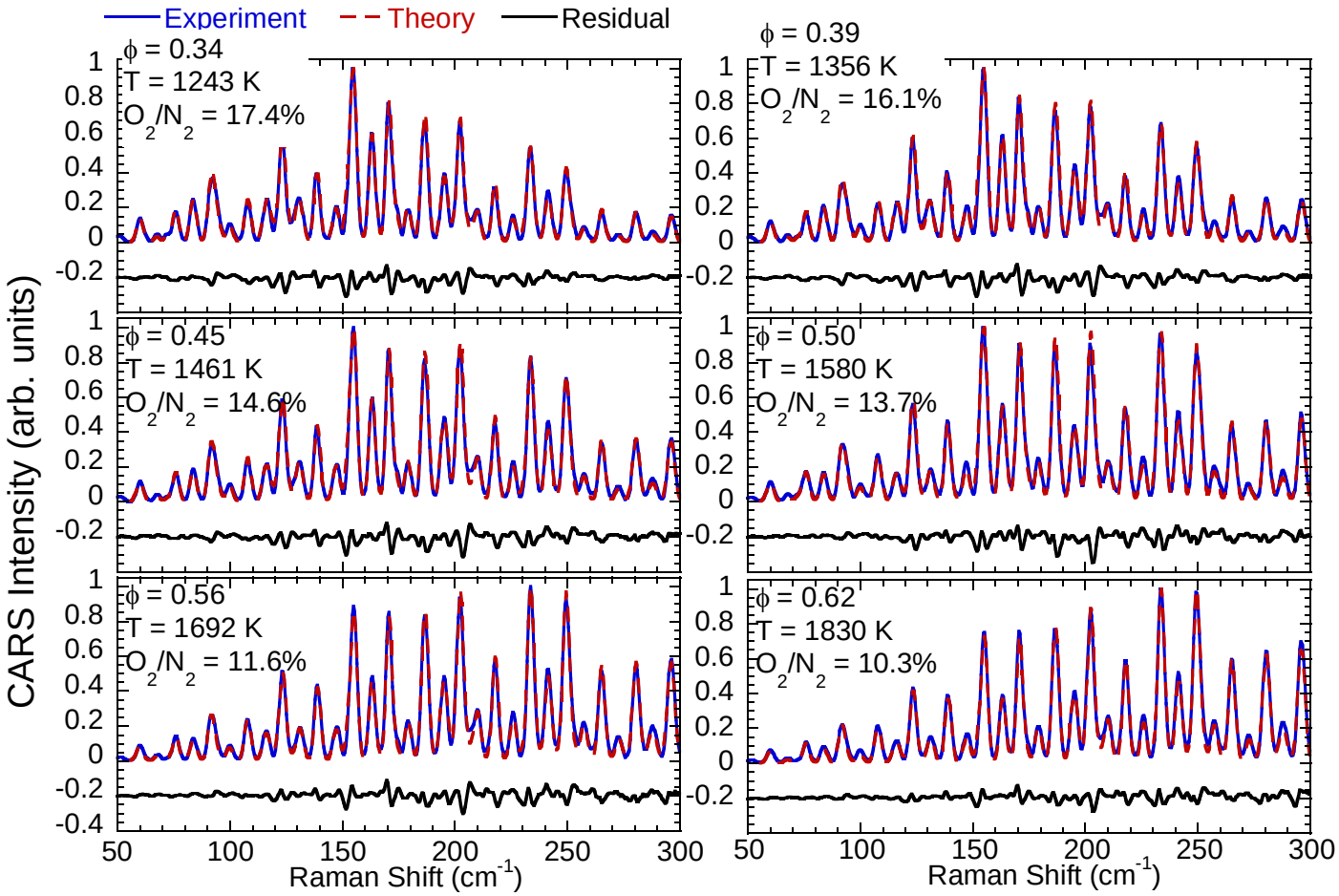


Temperature Field Measured by Rayleigh Scattering (CH_4/air)



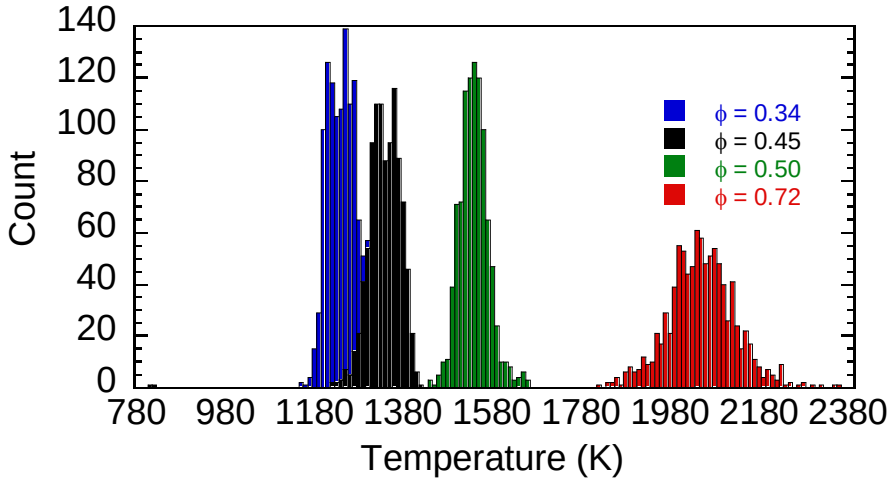
Burner Hole Configuration

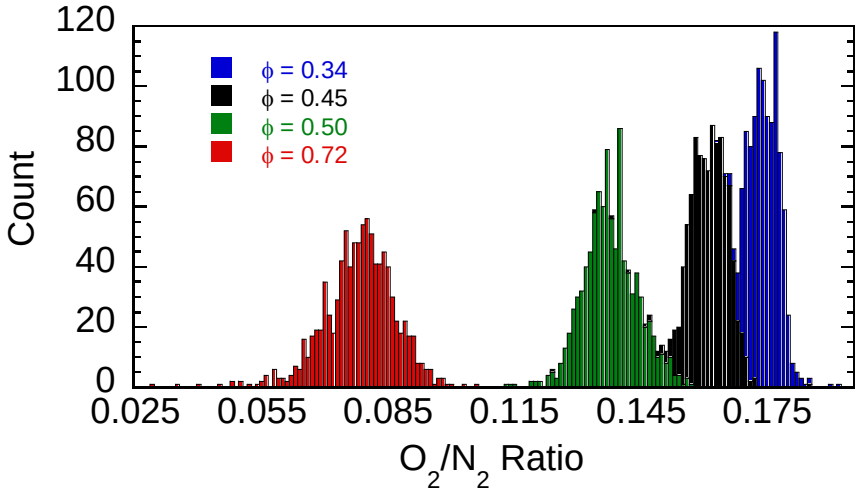
Shot-Averaged Spectra from Near-Adiabatic H_2/air flame



- Spectra averaged for several thousand laser shots to optimize SNR
- N_2 contributions dominate all spectra
- O_2 sensitivity arises from alteration of spectral "envelope" and subtle line shifts

Temperature and O_2/N_2 Measurement Precision



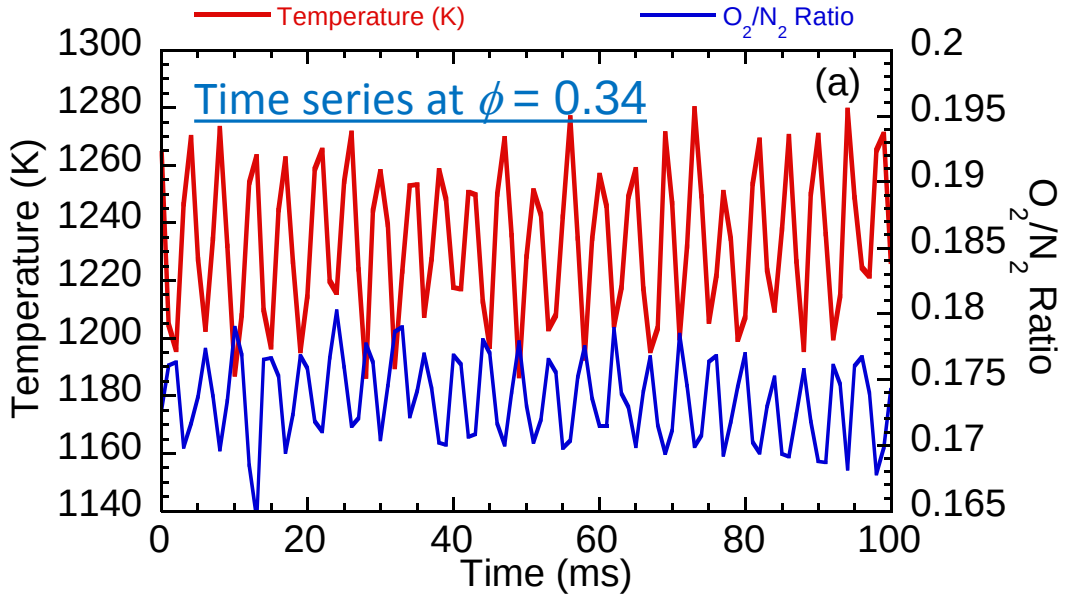


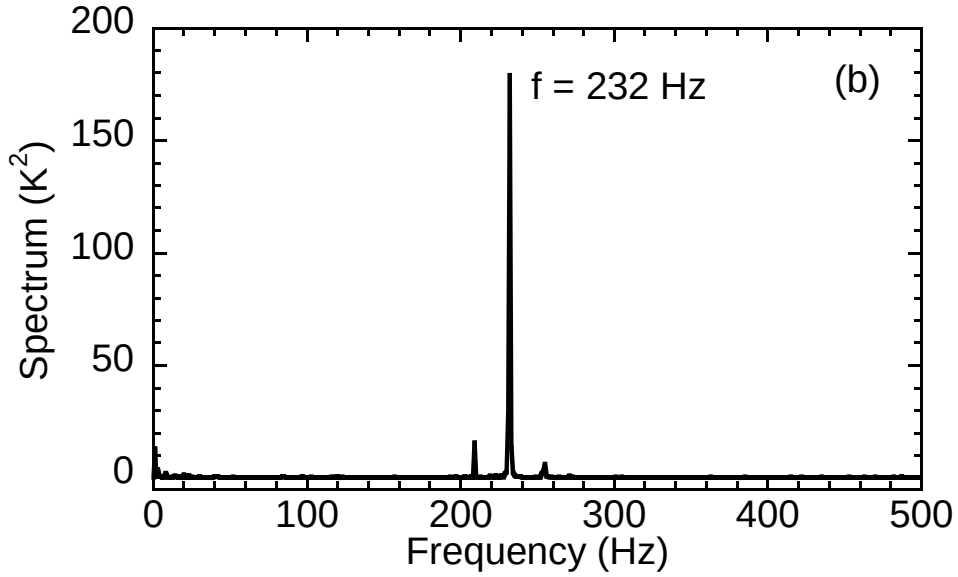
ϕ	T(K)	σ_T (K/%)	O_2/N_2	σ_{O2} (–/%)
0.34	1235	26.8/2.2%	17.4%	0.34/1.95%
0.39	1338	40.7/3.0%	16.1%	0.46/2.85%
0.50	1539	32.8/2.1%	13.7%	0.64/4.67%
0.72	2051	75.2/3.6%	7.8%	0.85/10.9%

EM Gain ON

- Temperature precision is 2-3% with EM gain off
- Best results with fs CARS are ~1%.
- ns rotational CARS ~ 3-4%

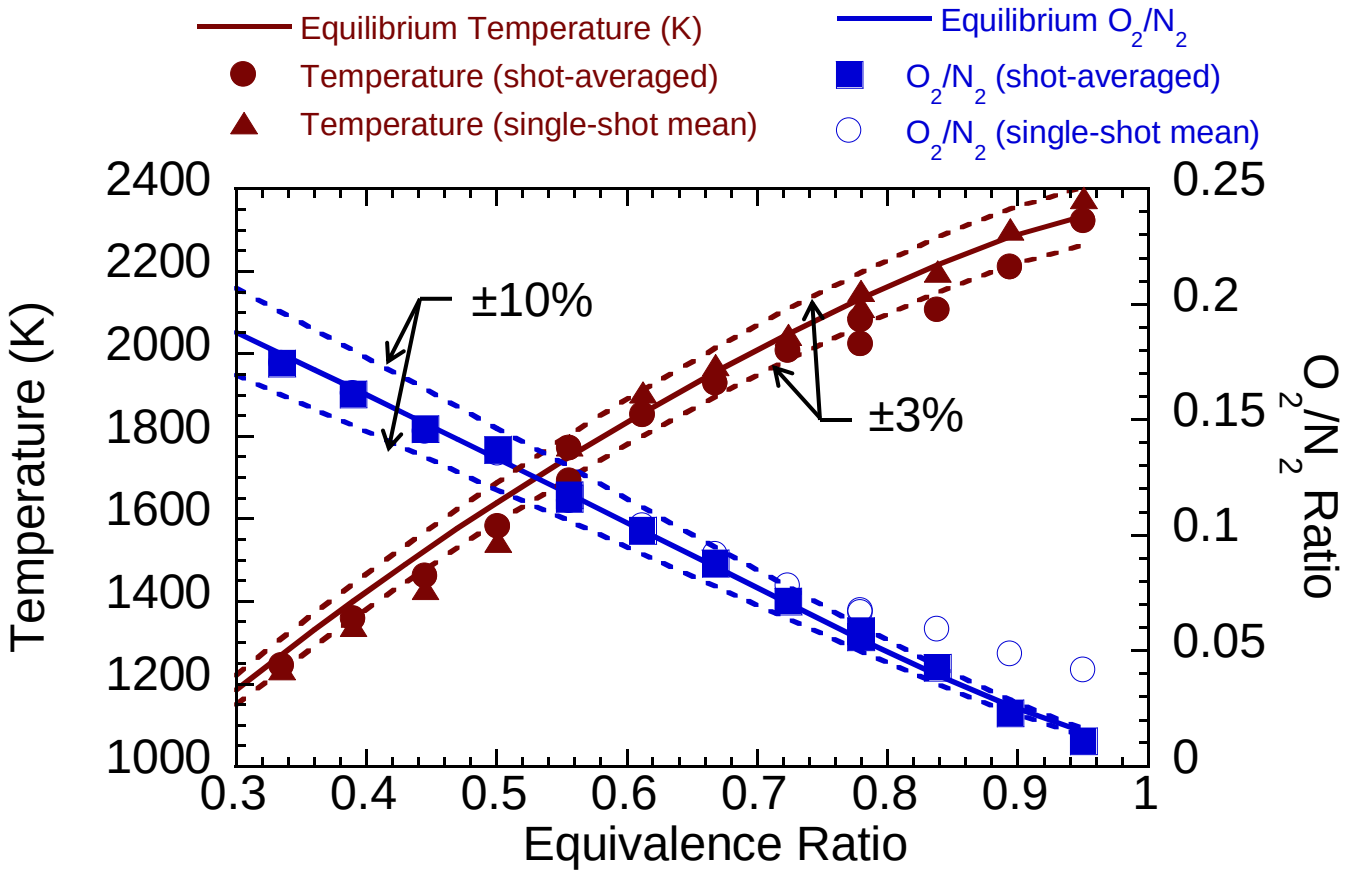
Acoustic Interaction at ϕ up to 0.55





- Audible "hum" from burner at leanest stoichiometries
- Inspection of T and O_2 data reveals oscillation in both signals
- Sampled at 1 kHz
- Negative correlation is consistent with low-level oscillations in ϕ
- Amplitude of temperature oscillation is ~30 K (2.4%)
- Distinct peak in PSD near 232 Hz is consistent with tone heard from the burner
- Precision could be understated

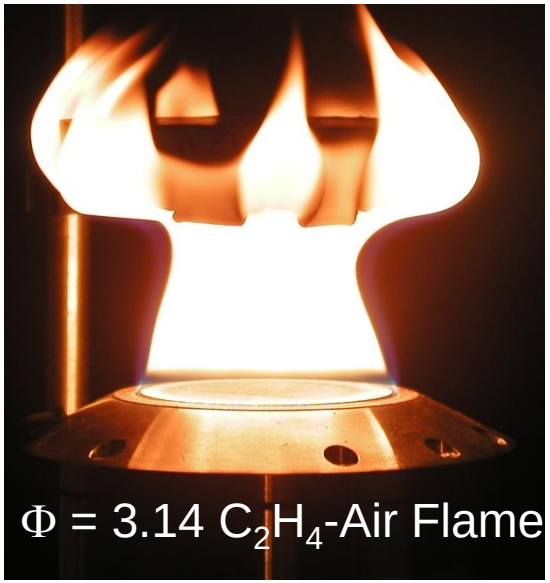
H_2/Air Flame Measurement Accuracy

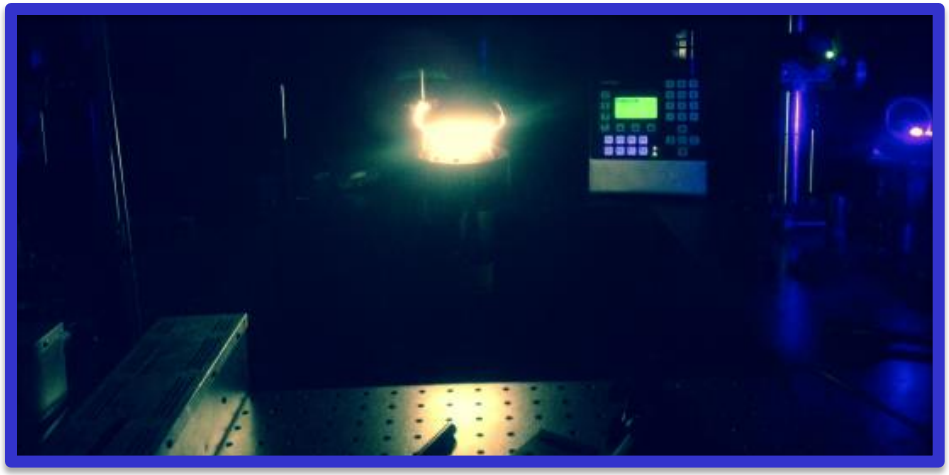


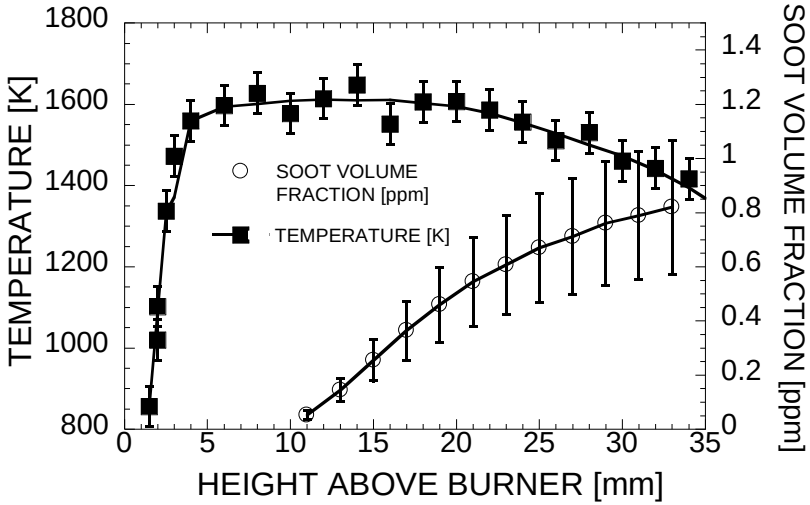
- Temperature accuracy: −3% when shot-averaged spectra are used
- Temperature accuracy: −3-6% when single-shot means are used
- O_2/N_2 accuracy is ±6%
- Uncertainty due to metered gas flows: Temperature ±3% / O_2/N_2 ±6-12%

C_2H_4/Air Flames on McKenna Burner

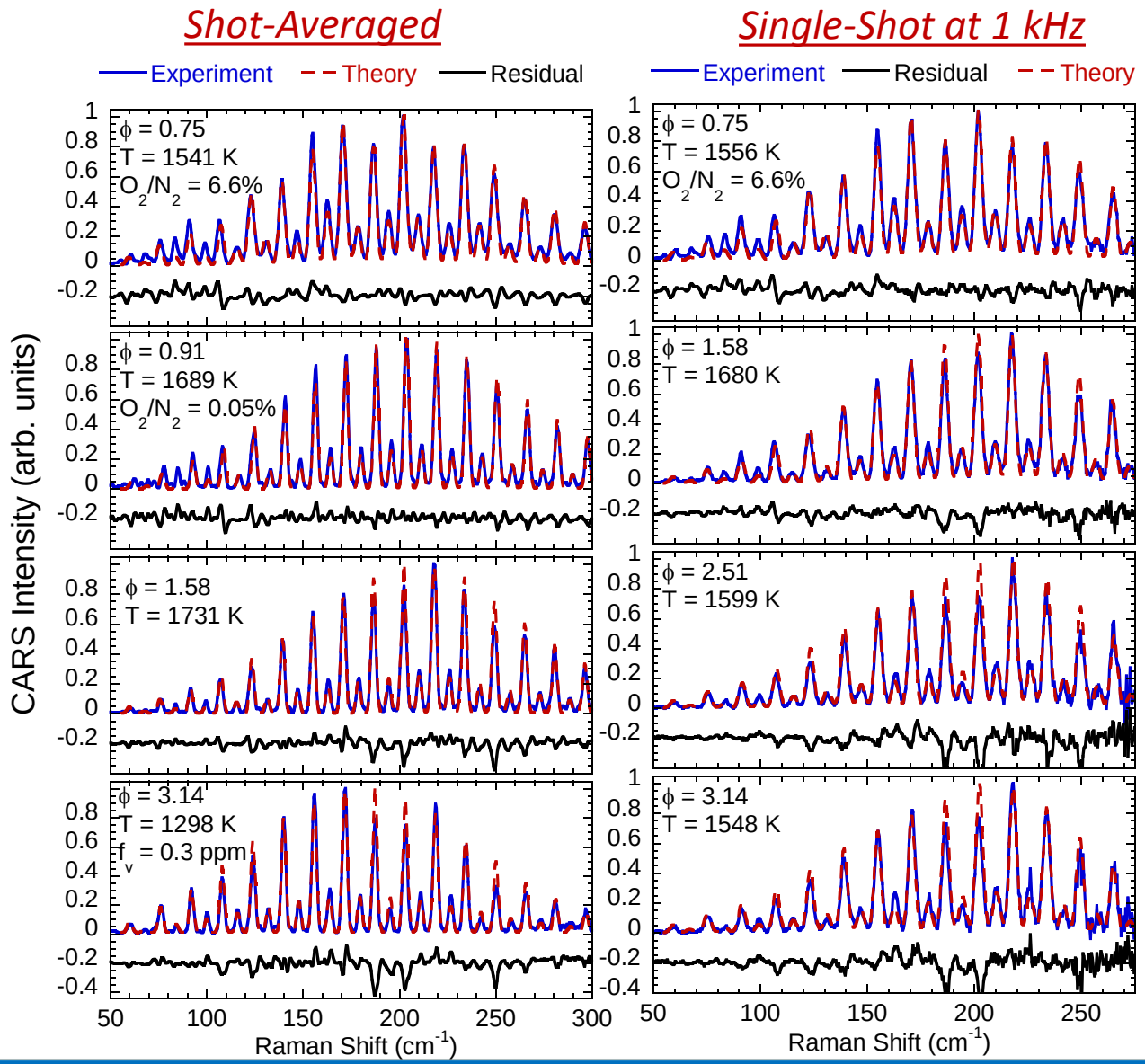
- Premixed hydrocarbon/air flame
- Water-cooled non-adiabatic burner
- Stable region ~5-15 mm above burner
- Previously studied at $\phi = 3.14$ in our lab (and elsewhere!)
- Wide range of stoichiometry, $\phi = 0.75$ to 3.14
- Potential contributions from N_2, O_2, CO, (CO_2 minimized by probe delay)





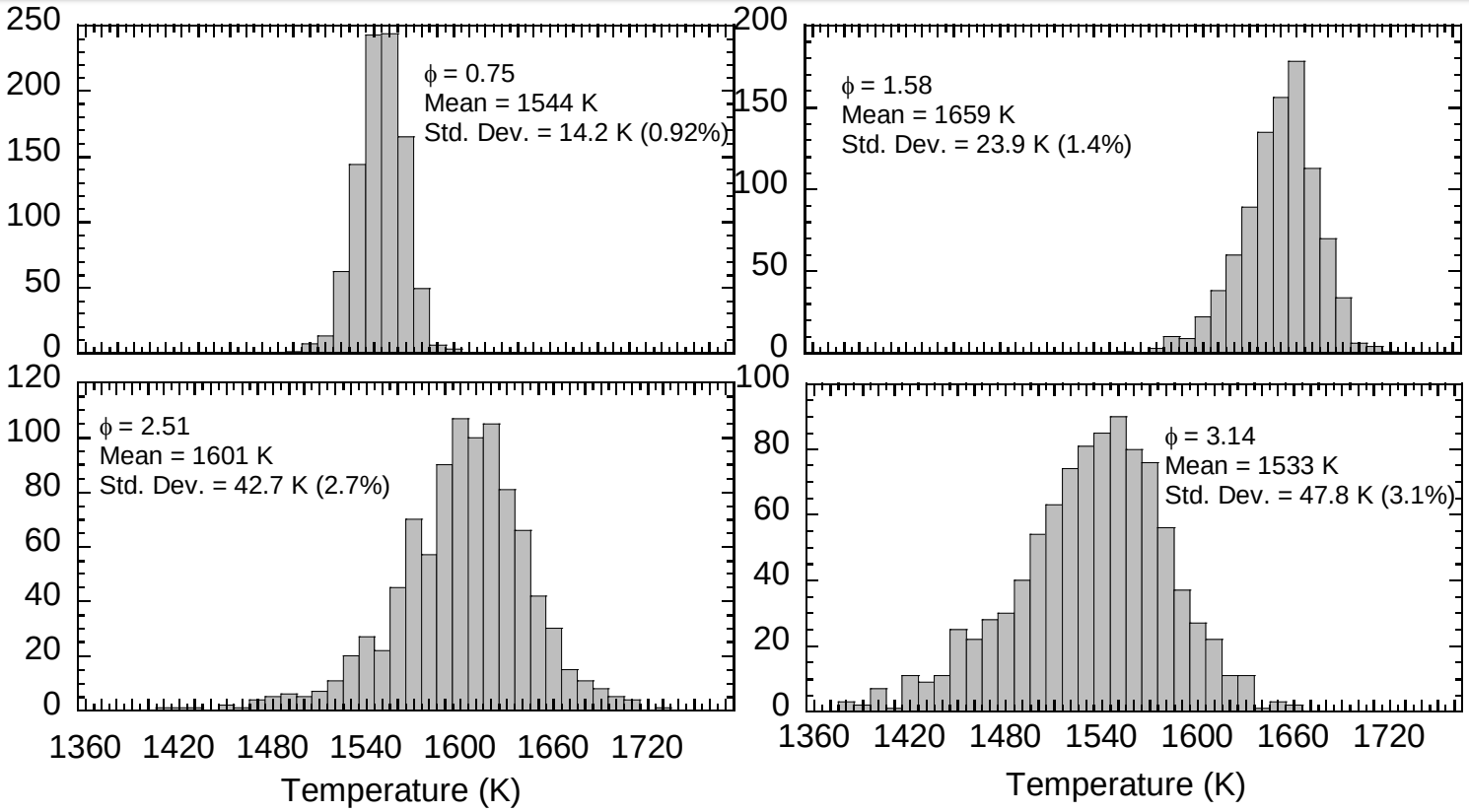


C_2H_4/Air Flame Spectra



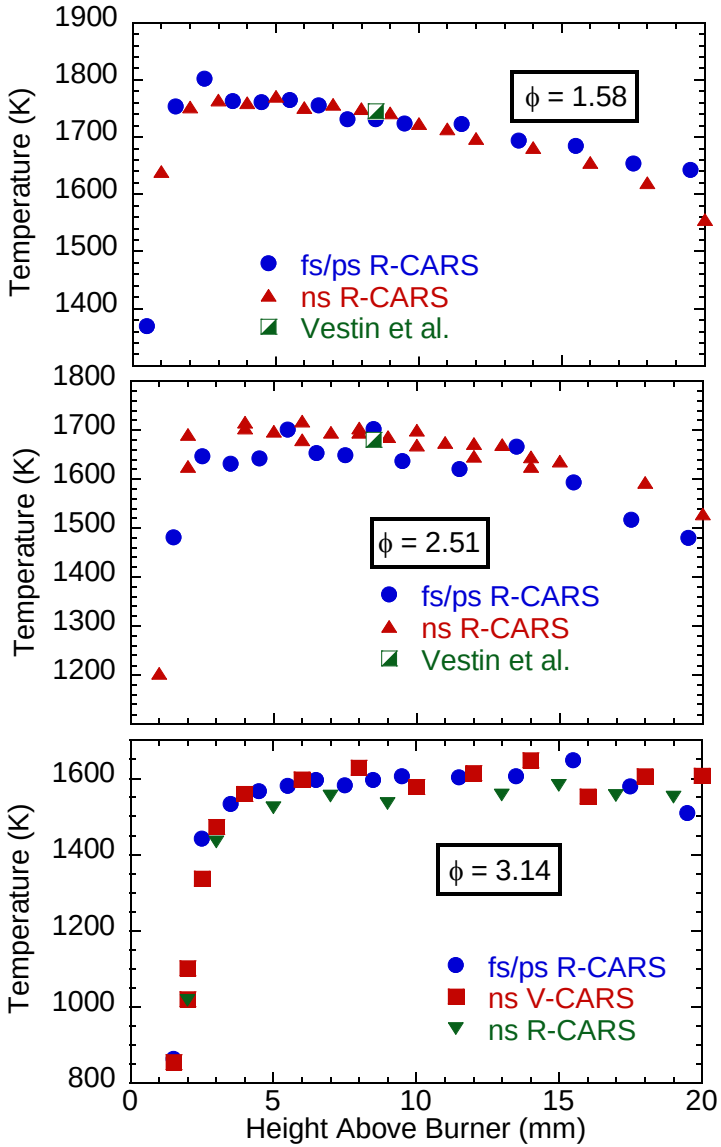
- Spectra acquired for fuel-lean to rich sooting flames
- High-quality fits observed for $\phi < 1$
- Systematic bias toward "underfit" of isolated lines for fuel-rich flames
- Fitted temperature appears to be robust
- Reliable spectra obtained in ***sooting regions*** of the flame
- ***No EM gain used***

Temperature Histograms C_2H_4/Air Flame



- Measurement volume positioned 11.5 mm above burner ***where flame is stable***
- Minimal variation in temperature in this range of ϕ
- Precision is 0.9 to 1.4% in leanest flames investigated : ***CARS photon yields are highest***
- Precision degrades to ~3% in richest flames
- Precision appears to be correlated to photon yield

Temperature Accuracy in C_2H_4/Air Flames: comparison to ns-CARS



- Comparison is most valid in stable region 5-15 mm height
- ns-CARS temperatures based on fits to shot-averaged spectra
- Agreement of fs/ps and ns CARS is within 1-3% in these ***fuel-rich, sooting*** flames
- Within reported accuracy of rotational ns-CARS
- ns-CARS measurements
  - Rotational CARS at Sandia 11/2013
  - Vibrational CARS at Sandia (2006)
  - Rotational CARS at Lund (Vestin et al. 2005)

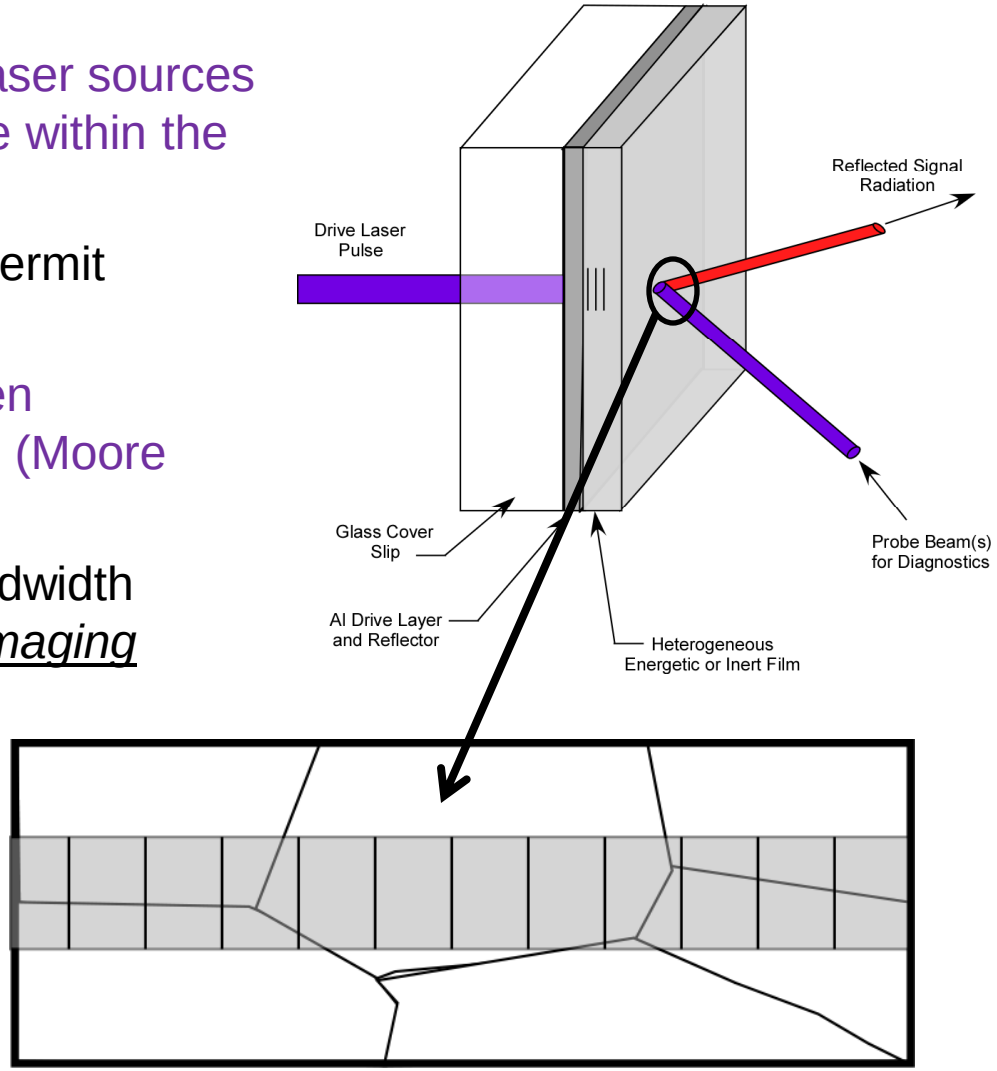
Summary and Conclusions

Sandia National Laboratories

- High-energy probes generated via SHBC enable flame temperature measurements with rotational fs/ps CARS
- We have performed a ***systematic assessment of accuracy and precision***
  - Near-adiabatic H_2/air flames
  - Premixed hydrocarbon air flames for ϕ = 0.75 to 3.14
- Temperature accuracy is 3% for shot-averaged spectra, 3-6% for single-shot means
- Uncertainty in temperature standards is 3%
- Accuracy of O_2/N_2 ratio is 2-6% in H_2/air flames
- Temperature precision is outstanding!
  - As good as 1%
  - Periodic oscillations of 30 K (2%) amplitude realized in Hencken burner
  - Results suggest correlation of CARS signal photons to precision
- Precision of O_2/N_2 ratio is 1-10% and monotonically increases with O_2/N_2
- O_2/N_2 sensitivity arises from spectral envelope and not resolution of O_2 lines

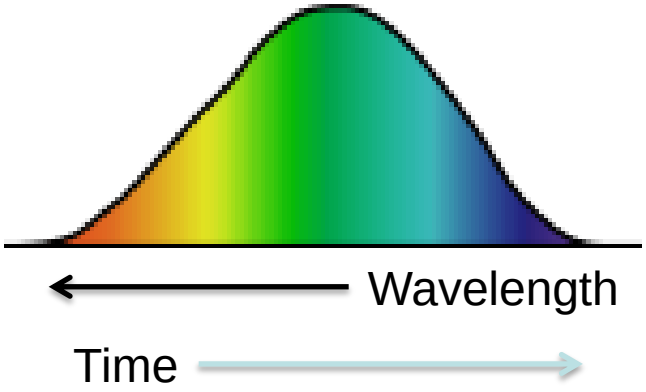
Our solution: Use femtosecond laser pulses for *mesoscale-resolved* experiments on *heterogeneous* materials

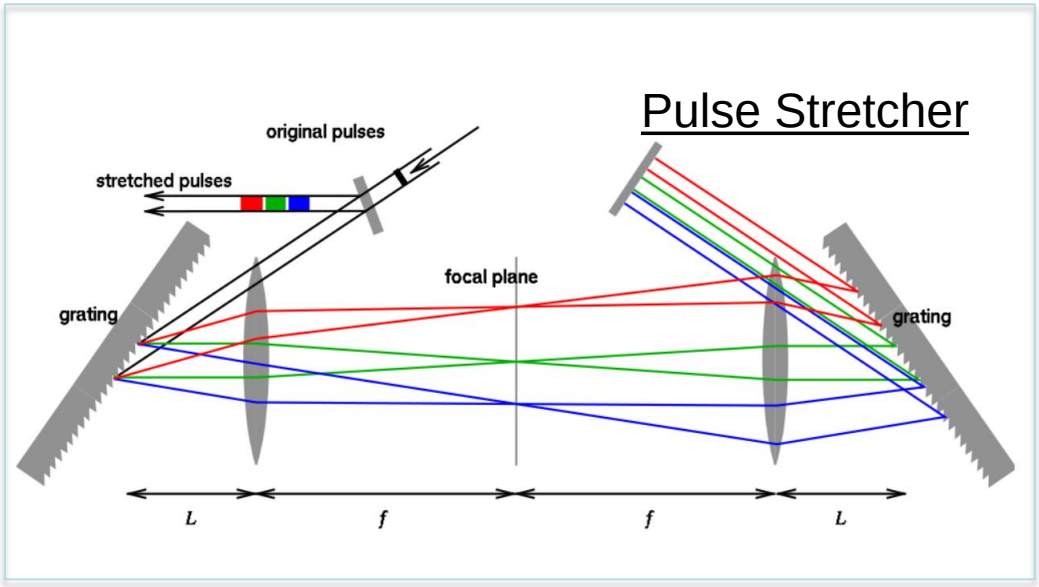
- Reliable femtosecond (fs) pulsed laser sources have become commercially available within the last ~10 years
- Ultrashort 10^{-14}–10^{-13} sec pulses permit access to ps-scale events
- Well-controlled laser drive has been demonstrated in homogeneous films (Moore group, LANL)
- Broadband yet low-noise laser bandwidth enables multiple spatially resolved *imaging* spectroscopies
- Spatially resolved measurements on *heterogeneous* samples at realistic grain sizes

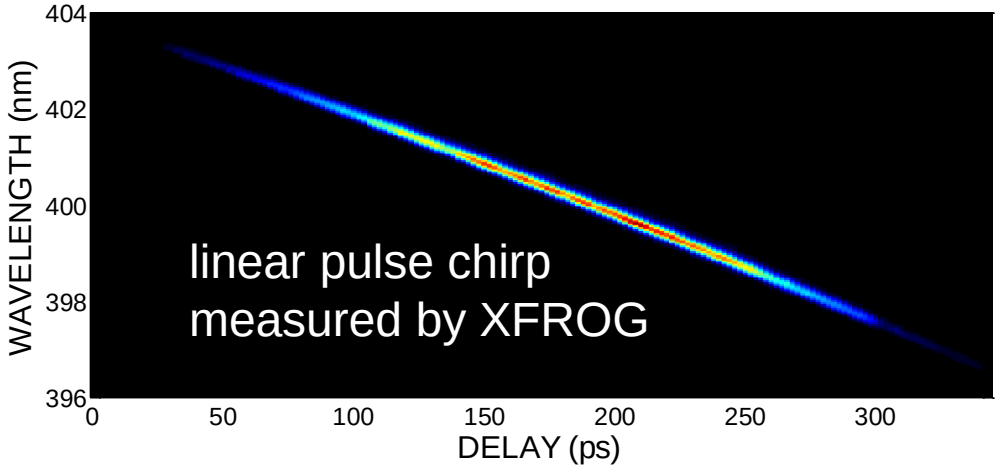


Femtosecond laser pulses have wide frequency spectra that allow us to encode time into frequency (color)

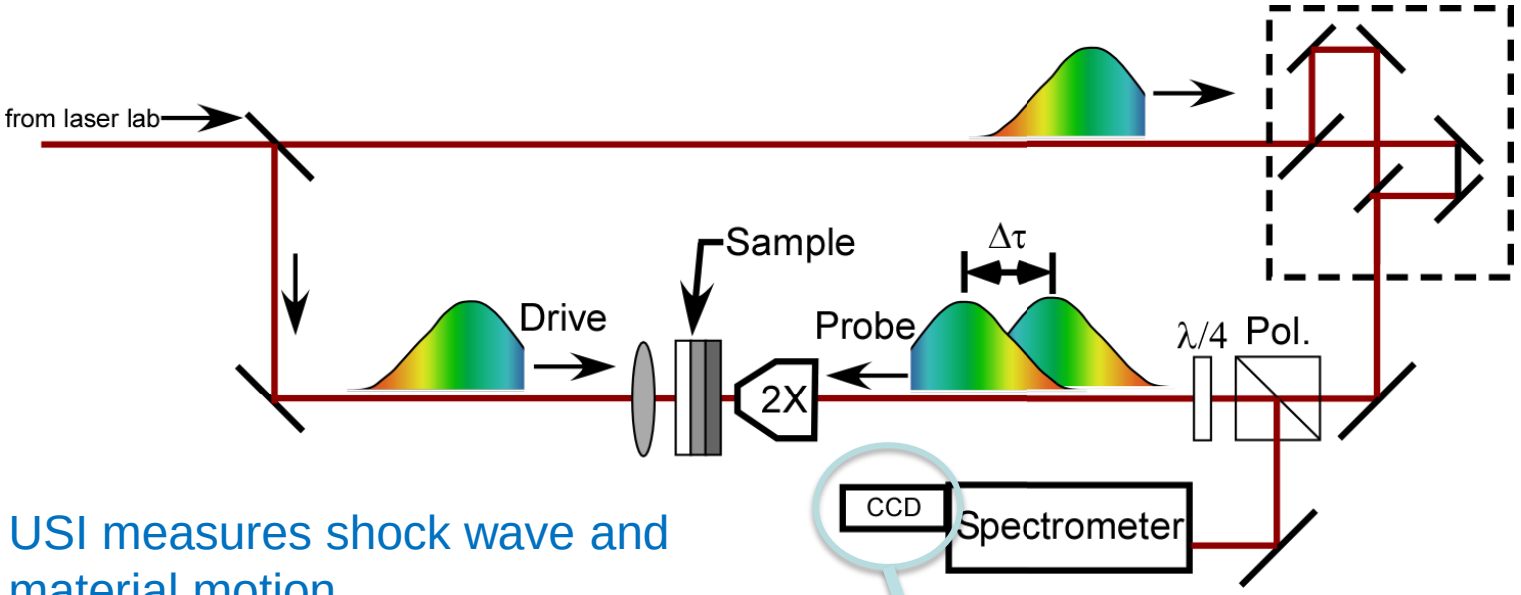
- Femtosecond bandwidths are typically 10s of nanometers
- Can be readily compressed or stretched (chirped) using grating-based stretchers
- Linear chirp is added in our lab to **encode wavelength into time with picosecond resolution**



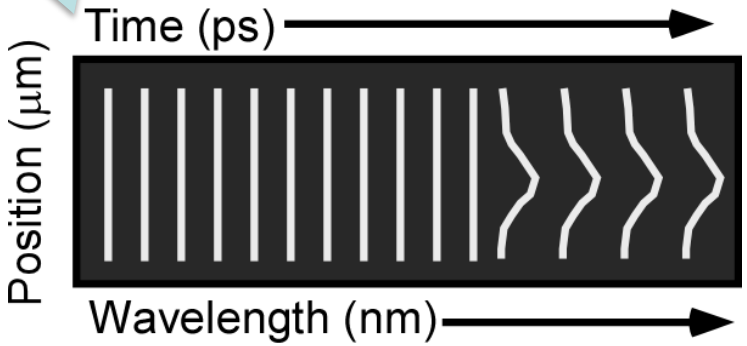




Ultrafast Shock Interferometry (USI) for characterization of shockwave structure and EOS measurements

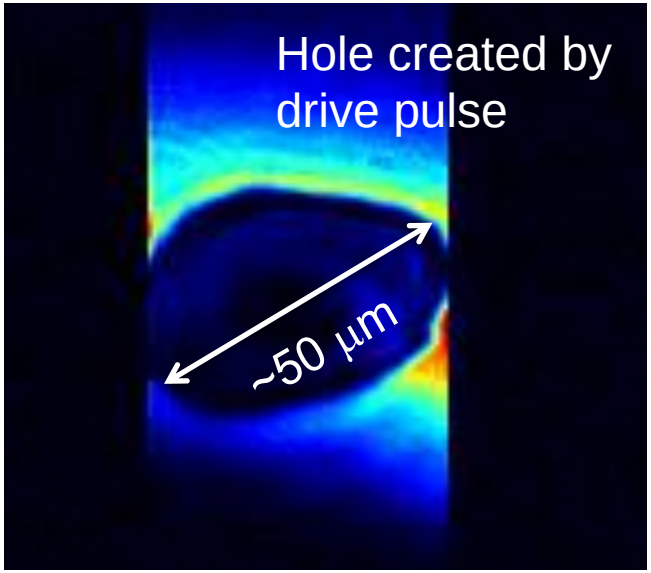


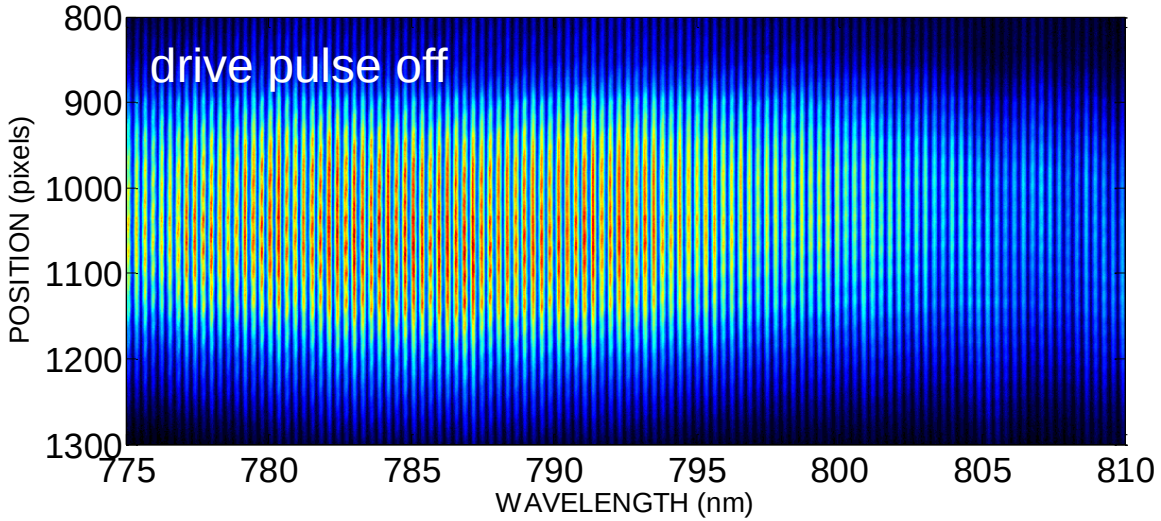
- USI measures shock wave and material motion
- Picosecond time resolution
- Micron-scale spatial resolution
- Laser-driven shock supported for 300-400 ps
- Shock-induced phase shift accumulated during probe separation, $\Delta\tau$, is monitored

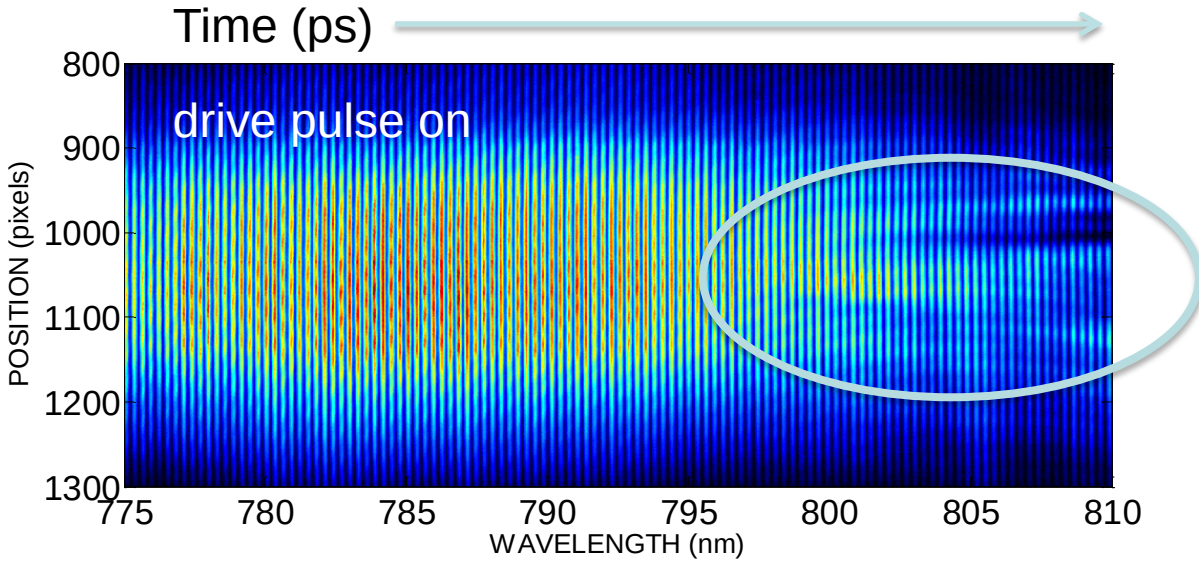


USI interferograms: laser-shocked aluminum films

- 1-micron Al film on glass substrate
- Ablator surface motion is tracked
- Surface velocity vs time and shock breakout profiles monitored







USI-measured ablator surface velocity histories