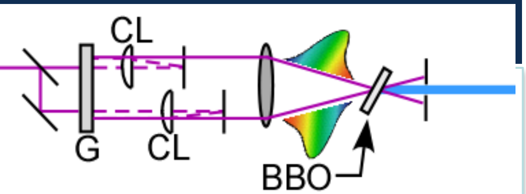
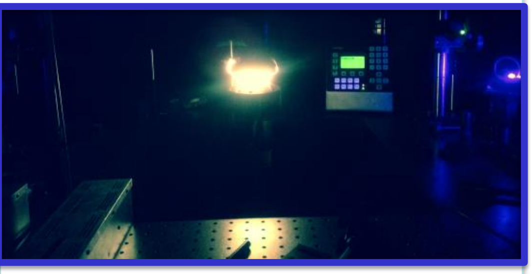
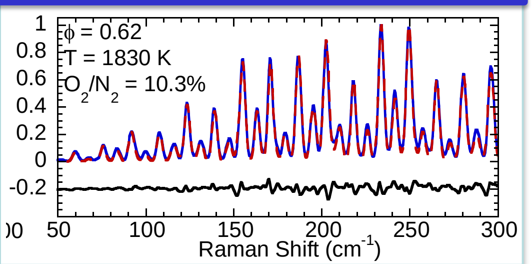
SAND2014-2238C







Femtosecond CARS for high-data rate, low noise temperature/species detection

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Sandia National Laboratories

Exceptional service in the national interest

U.S. DEPARTMENT OF ENERGY

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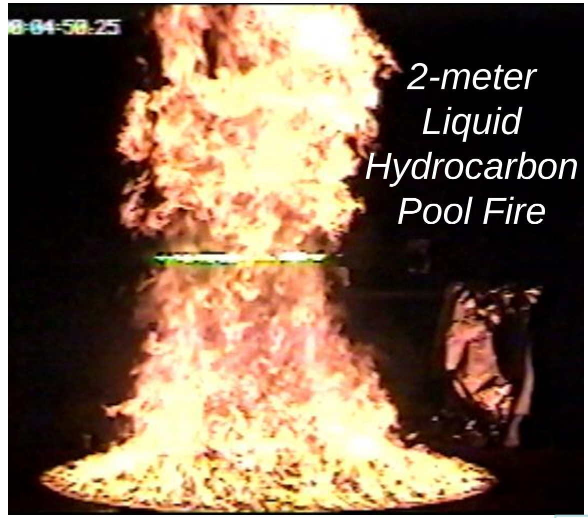
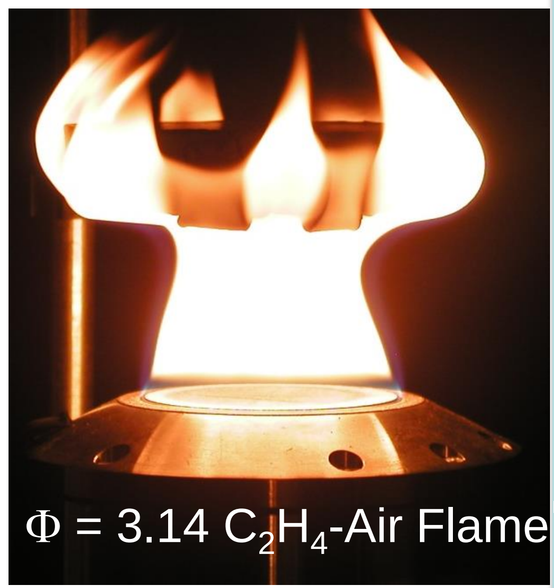
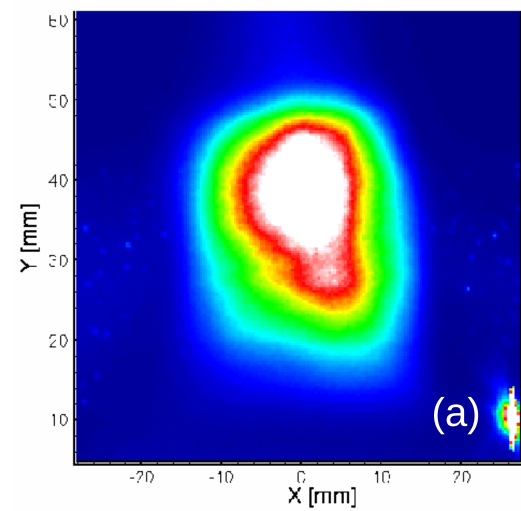
Sandia's application space presents significant challenges

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



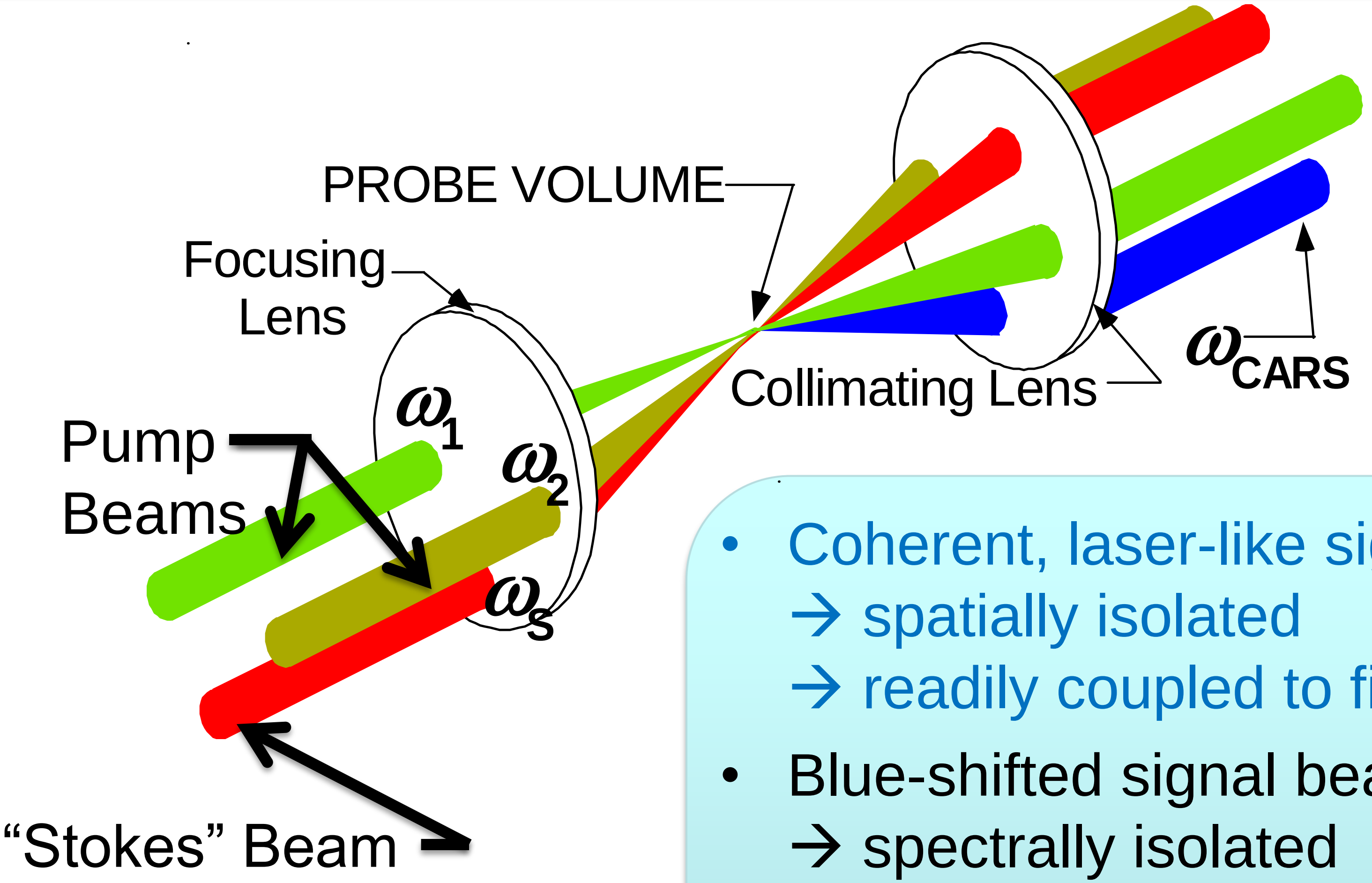
Propellant Burn





Sooting

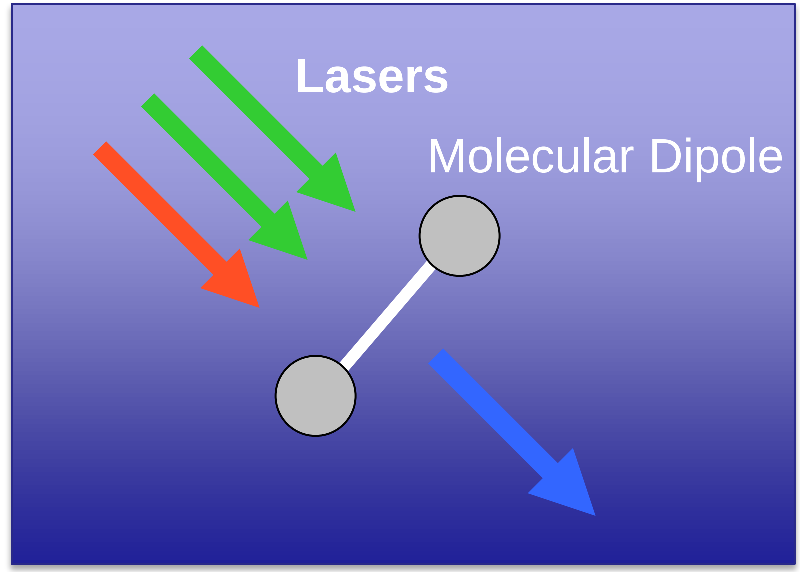
Coherent anti-Stokes Raman Scattering (CARS)



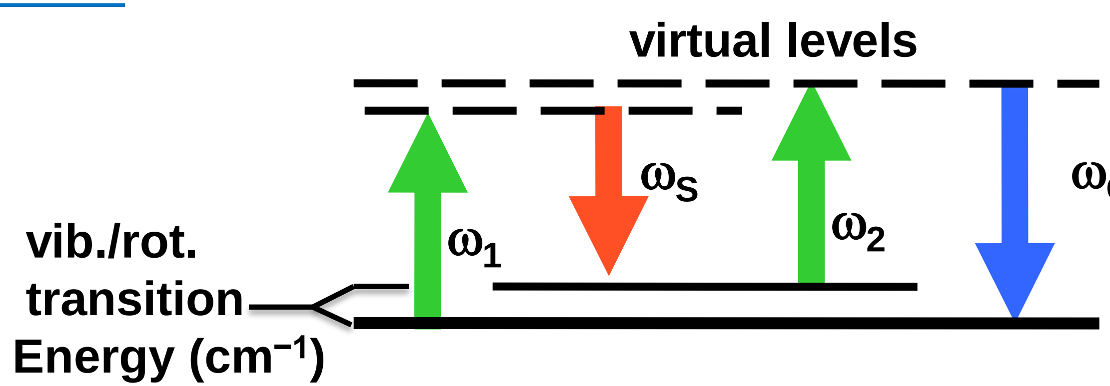
- Coherent, laser-like signal beam
  → spatially isolated
  → readily coupled to fibers
- Blue-shifted signal beam
  → spectrally isolated
- Orders of magnitude stronger than incoherent scattering

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

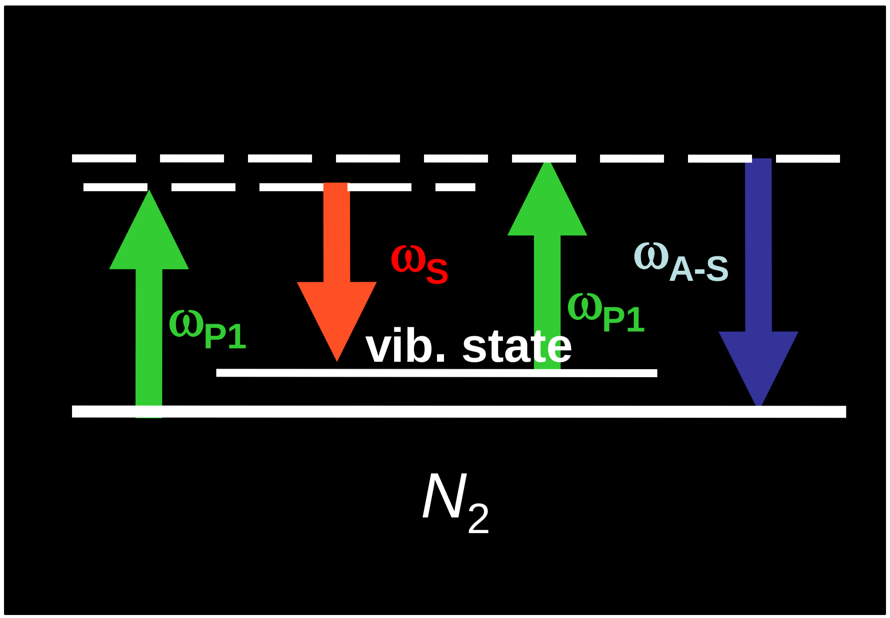


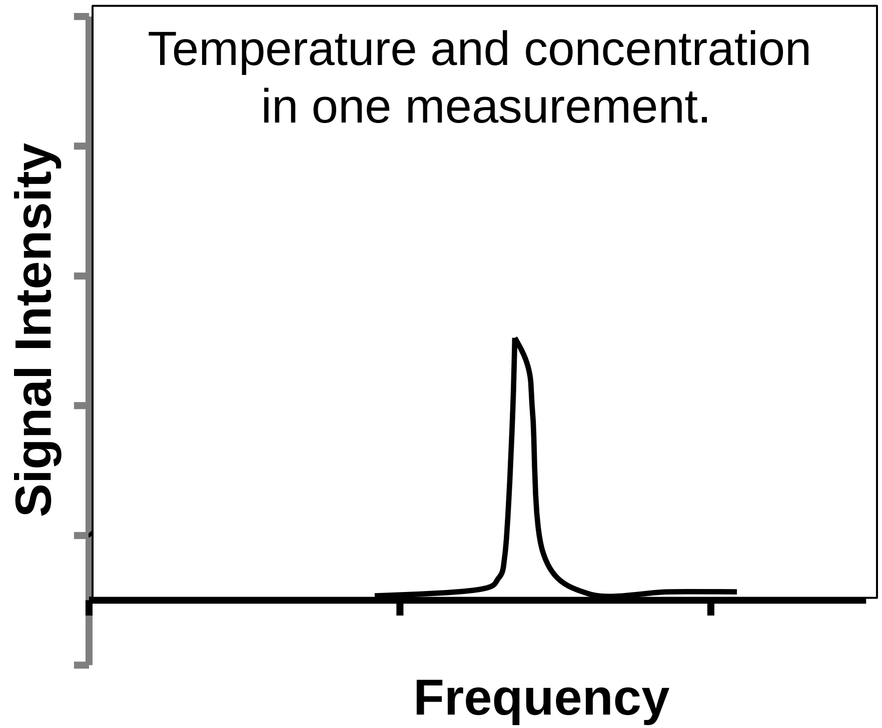
Coherent Anti-Stokes Raman



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

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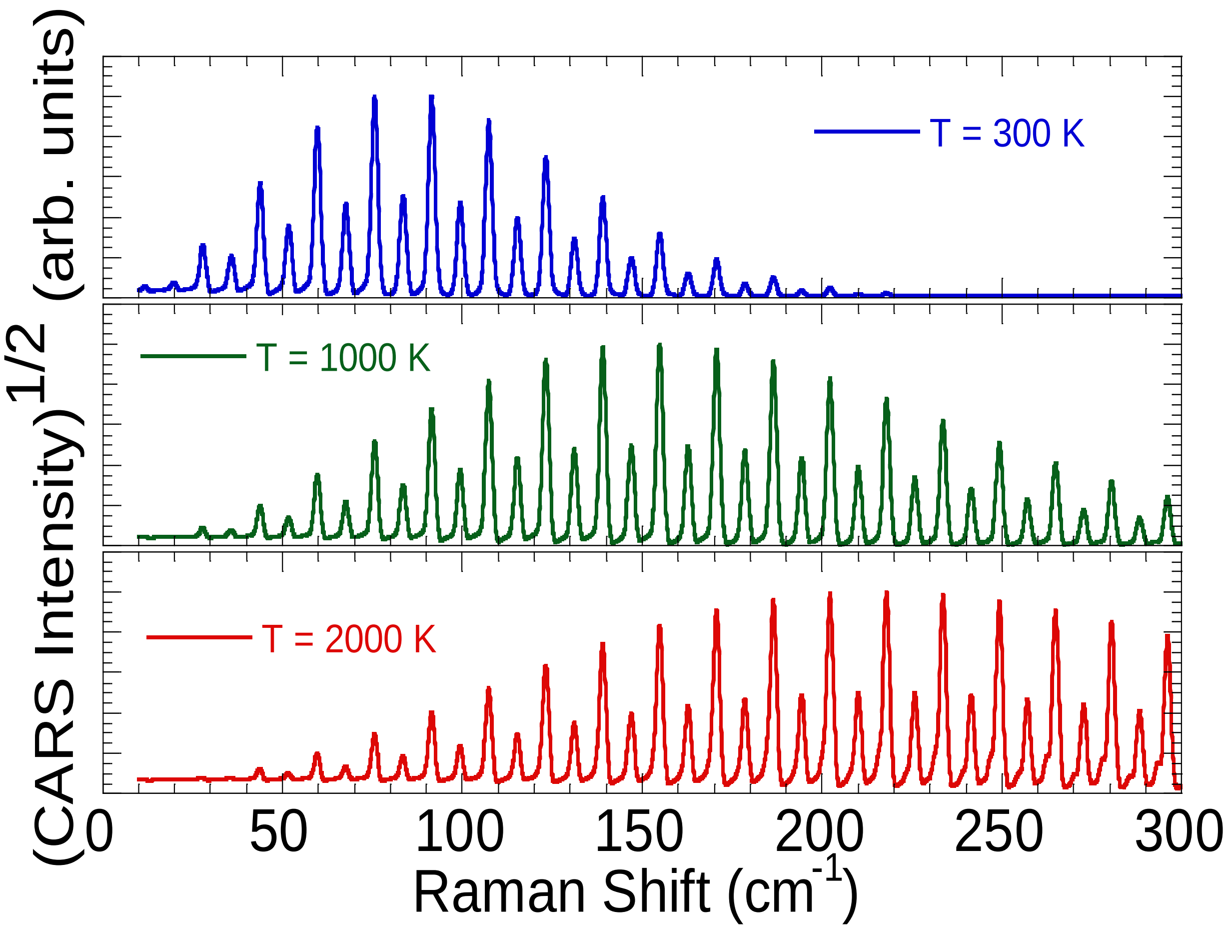


Model calculations are fit to experimental spectra

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

Boltzmann Fractions

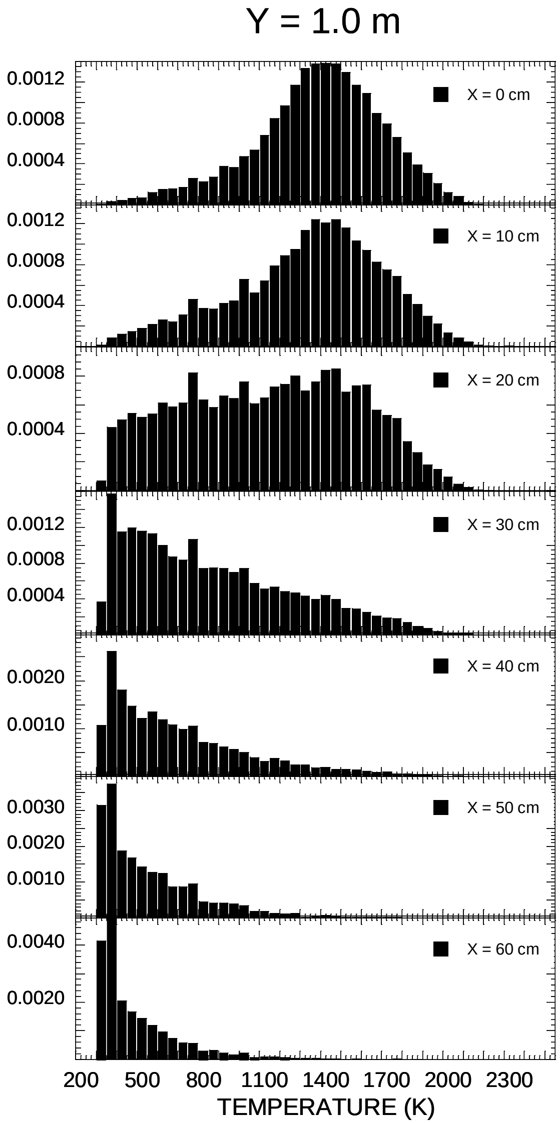
$$\frac{N_j}{N} = \frac{g_j \exp\left(-\varepsilon_j / kT\right)}{\sum g_j \exp\left(-\varepsilon_j / kT\right)}$$

Temperature dep.

Linewidths
Empirical!
Uncertain!

Successful CARS Application in Hostile Environment

- 2-m base diameter luminous sooting fire plume
- CARS temperature/species statistics
- Combined with LII for soot radiative emission statistics



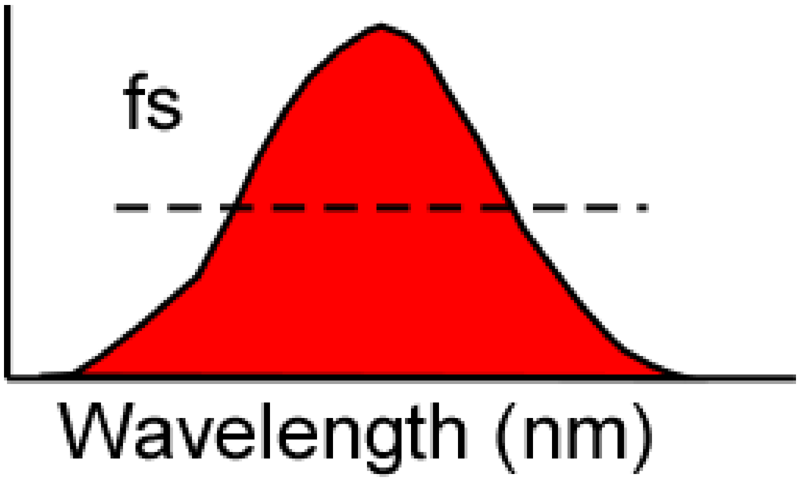
Why Ultrafast?

- High repetition rates (kHz vs 10 Hz)
- Collision free measurements! → no linewidths!

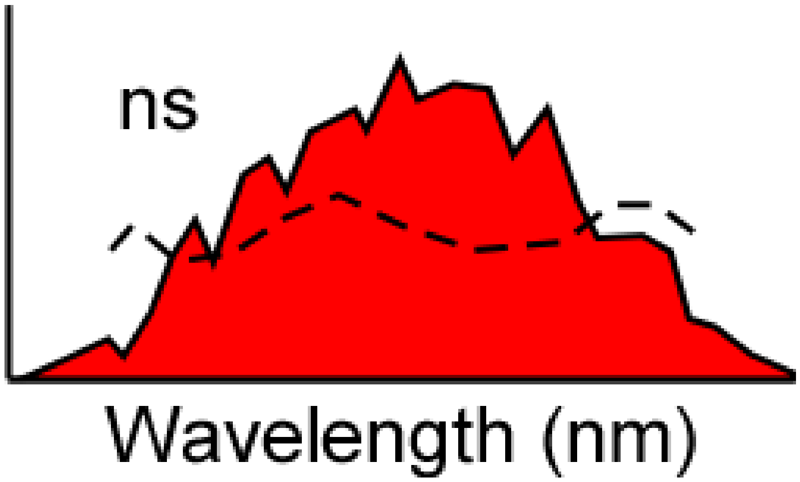
$$\tau_{laser} << \tau_{collision} \; \& \; \tau_{Raman}$$

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

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

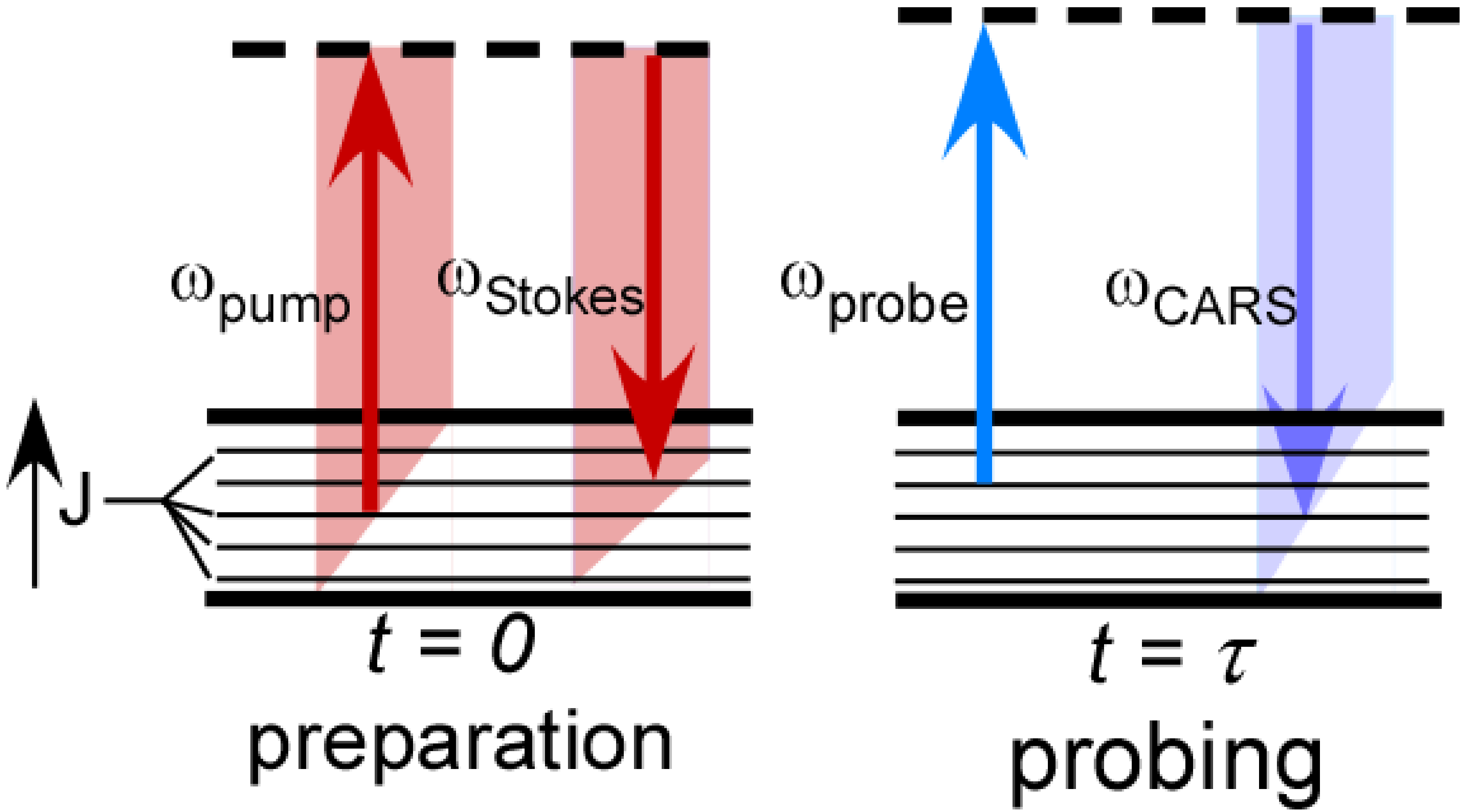


Transform limited



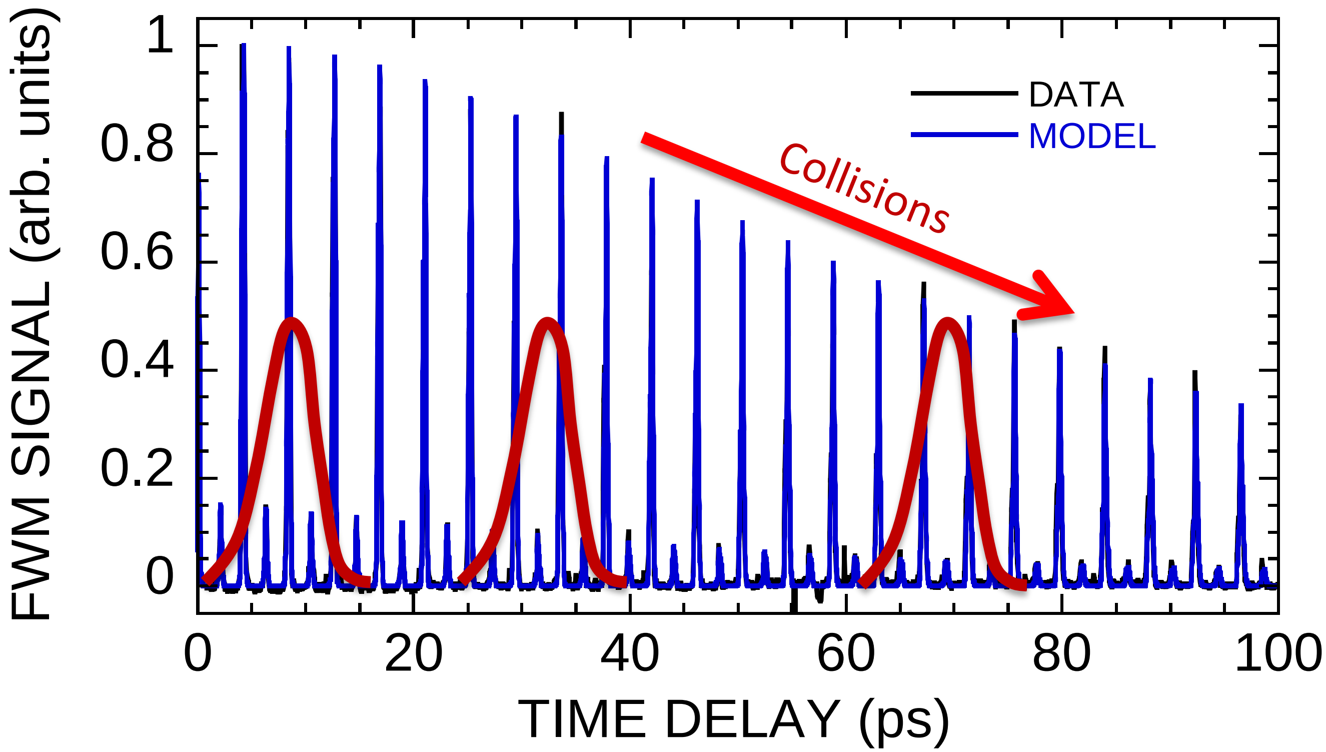
Chaotic!

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



"The story here is really in the time domain"

Measured Response in N_2 at T = 300 K



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

Proof-of-Concept Experiments in Air

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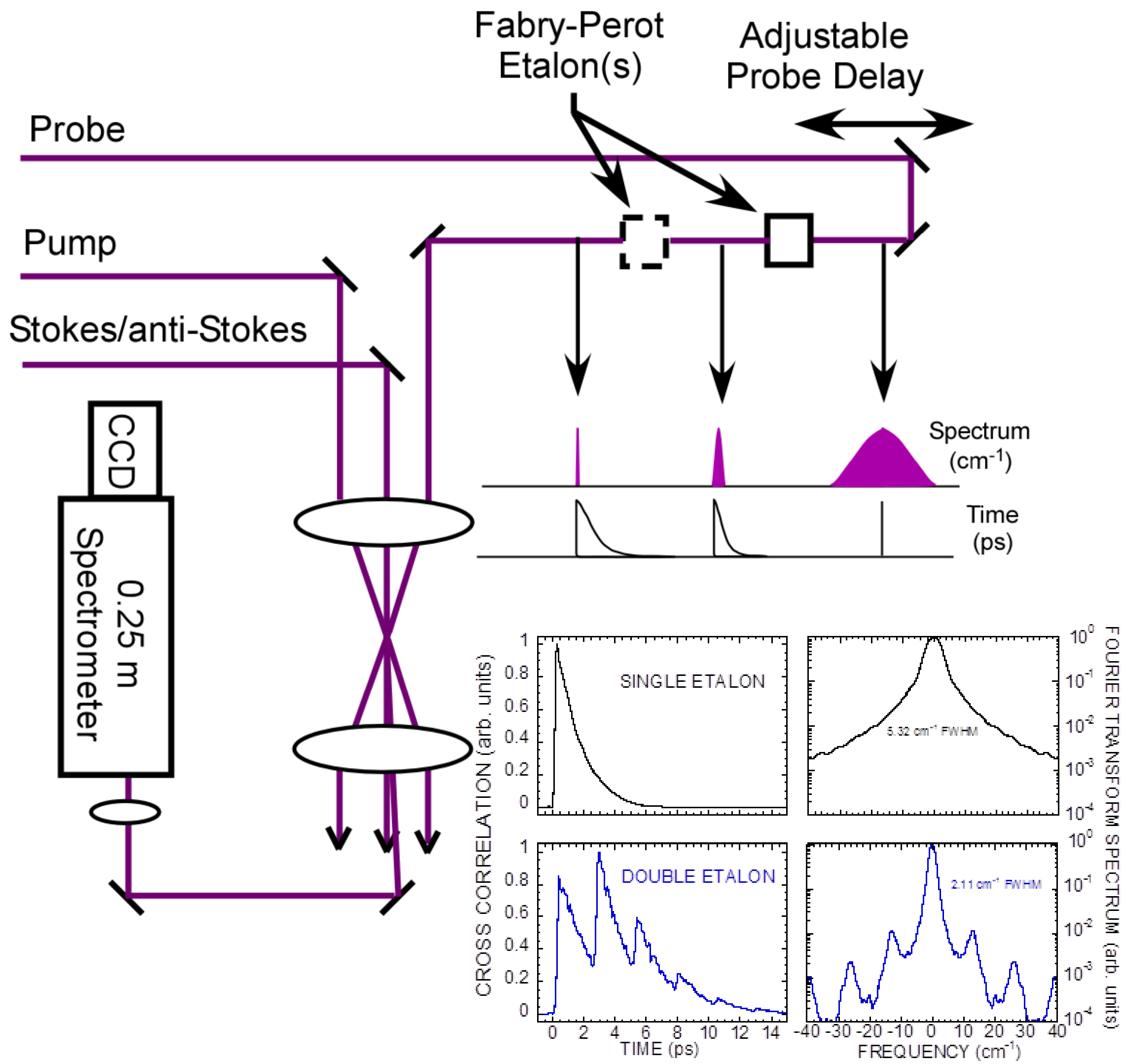
- **"Bandwidth-Carving"** to generate ps probe pulse
- Two different probe resolutions investigated with single- and double-etalon configurations
- Very inefficient (0.8 to 2.4% or less transmission)
- Atmospheric air spectra in tube-furnace up to 800 K for probe delays up to 20 ps



Measured and model-fit spectra in Air up to 800 K –1.5 ps probe

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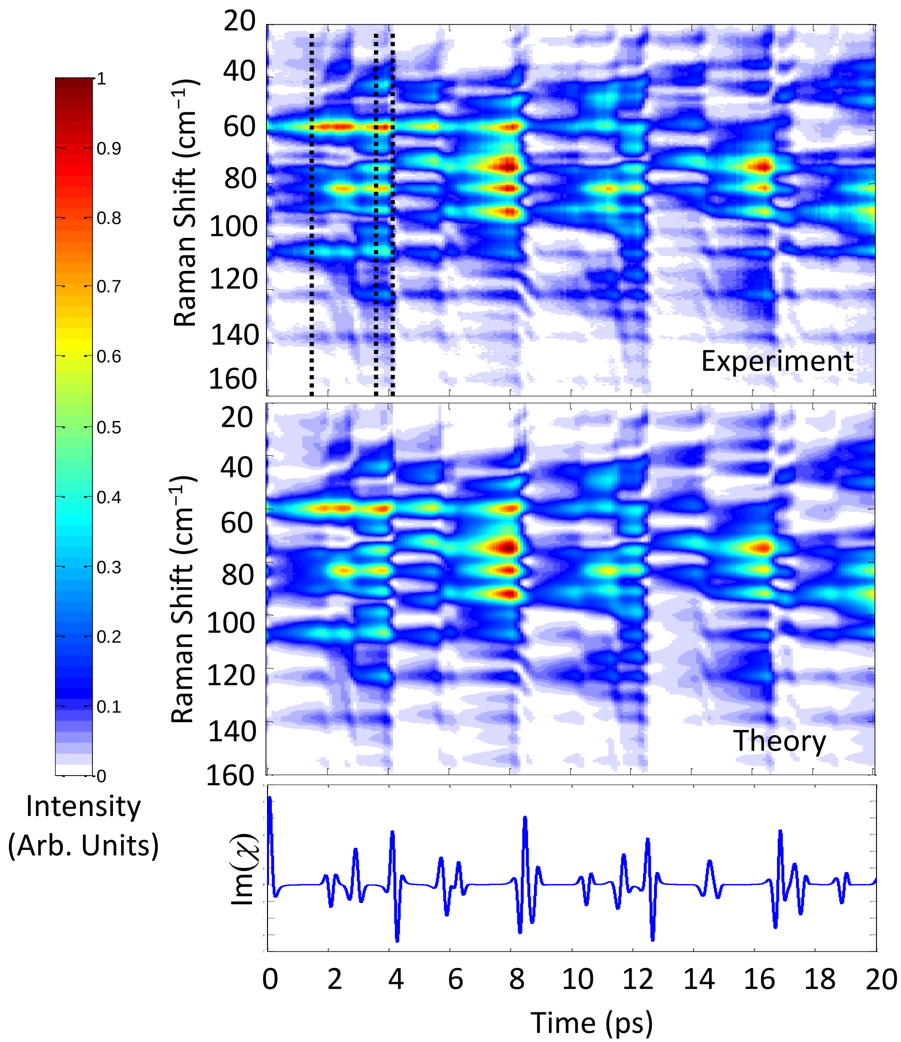
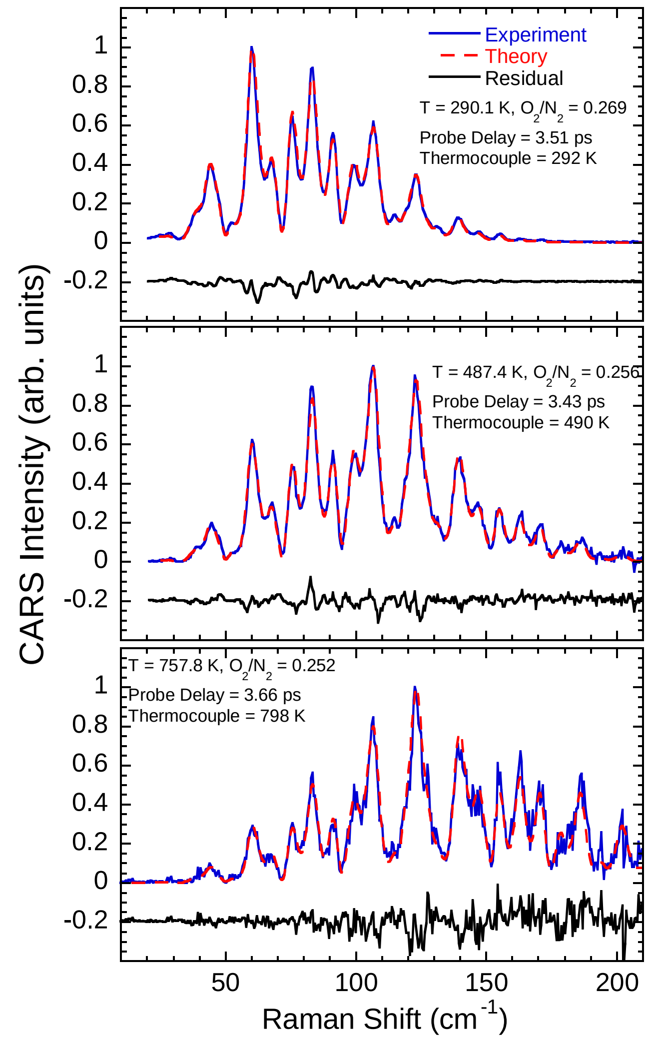
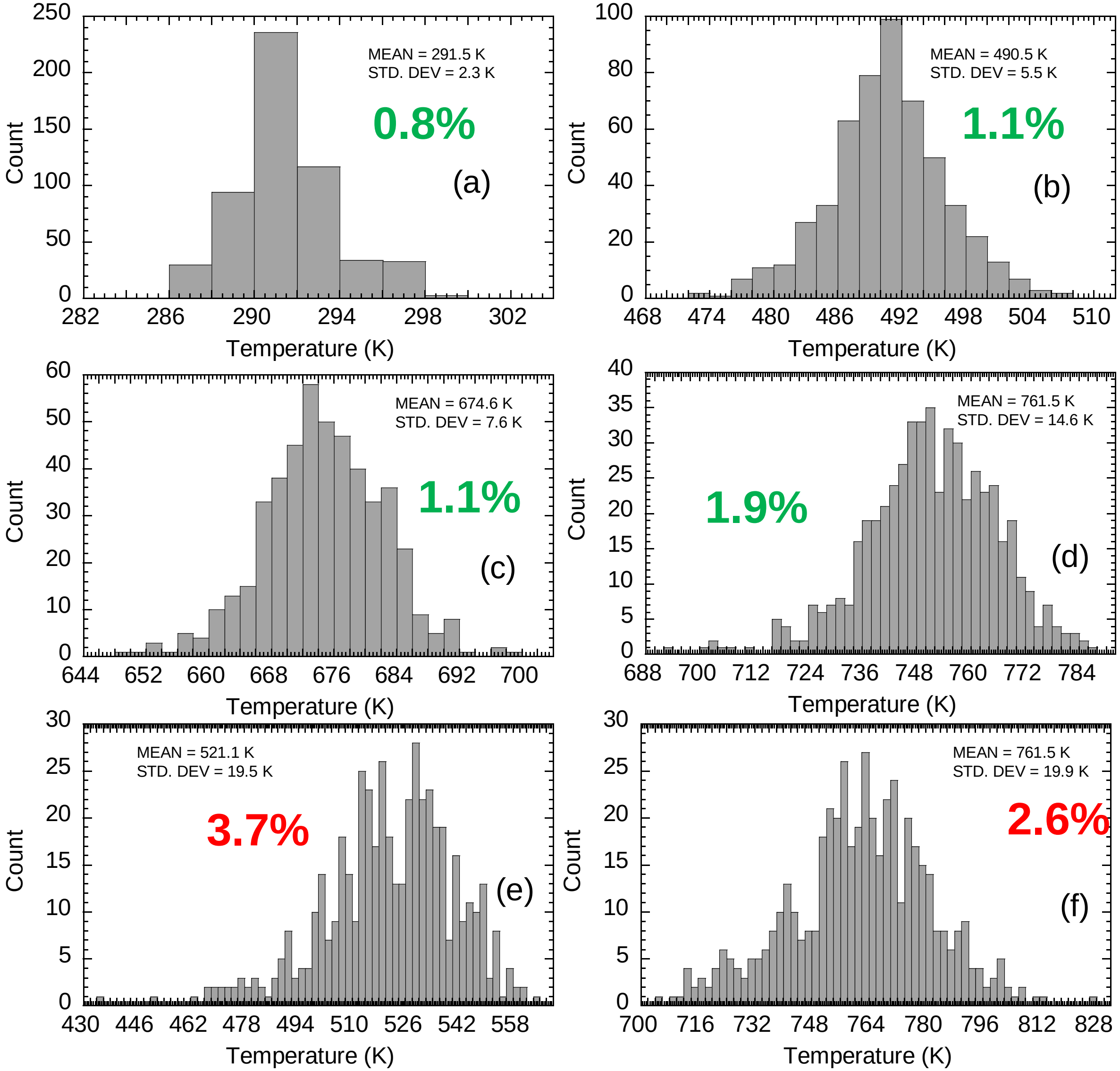


[Room temperature dynamics]



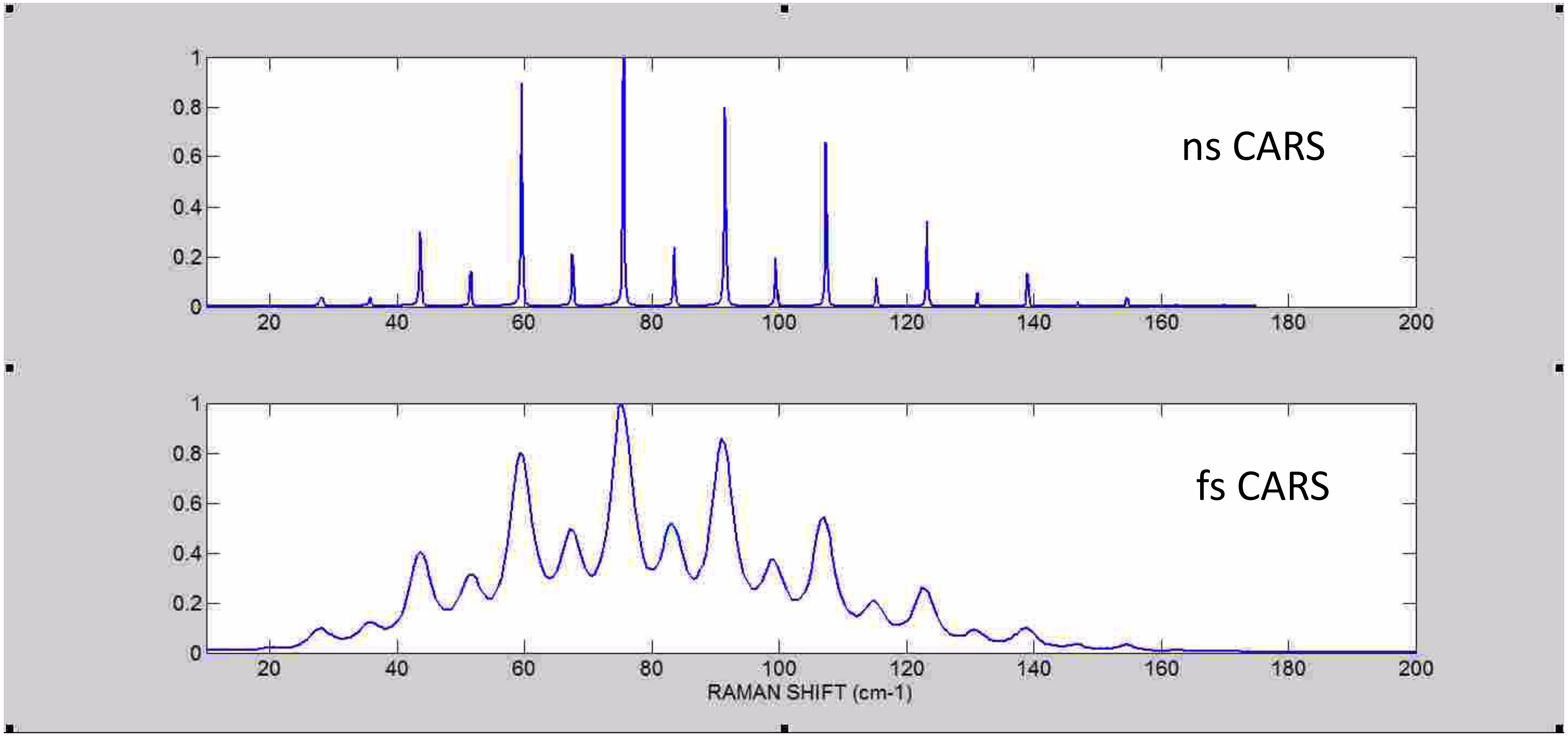
[Furnace temperature/O2 measurements]

Single-shot temperature histograms show high precision



- Histograms constructed from 1000 single-shot (1-sec) realizations
- Precision indicated on each plot
- Compare to ~4% typical precision with nanosecond sources
- Precision ~1% when broadband source is dominant noise source
- Precision degrades at lower photon yields when detection noise is dominant source

Low noise fs bandwidth can improve single-shot precision



Room-temperature N_2 spectra

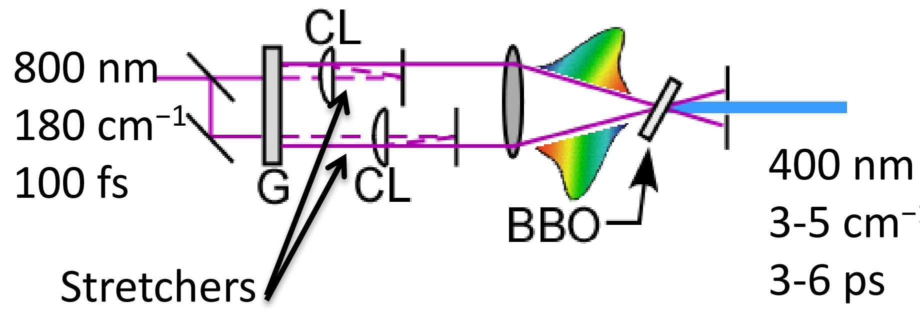
Second-Harmonic Bandwidth Compression (SHBC)

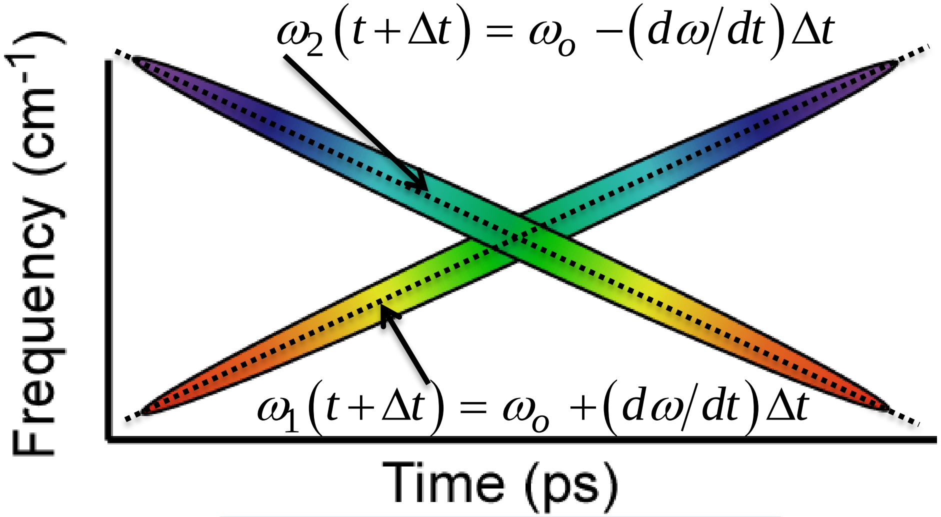
- Commercial device (Light Conversion)
- Converts fs radiation at 800 nm to ps radiation at 400 nm
- Grating pulse stretchers
- **<u>Phase-conjugate</u>** temporal chirps imparted upon broadband fs pumps
- Sum-frequency generation in BBO
- Output linewidth 3.5-4.0 cm^{-1}

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

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

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





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

Single-Laser Shot Spectra from Near-Adiabatic H_2/air flame

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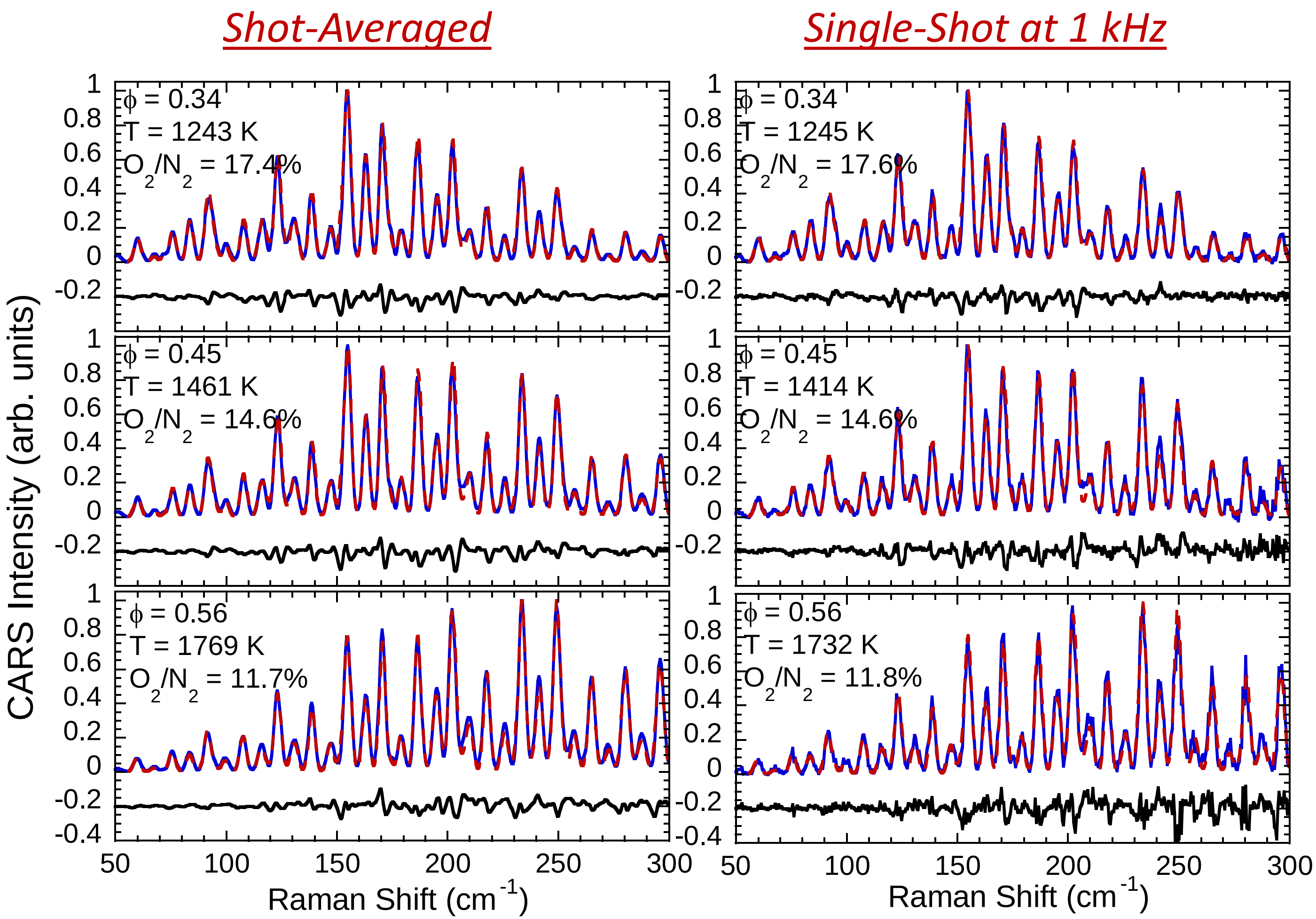
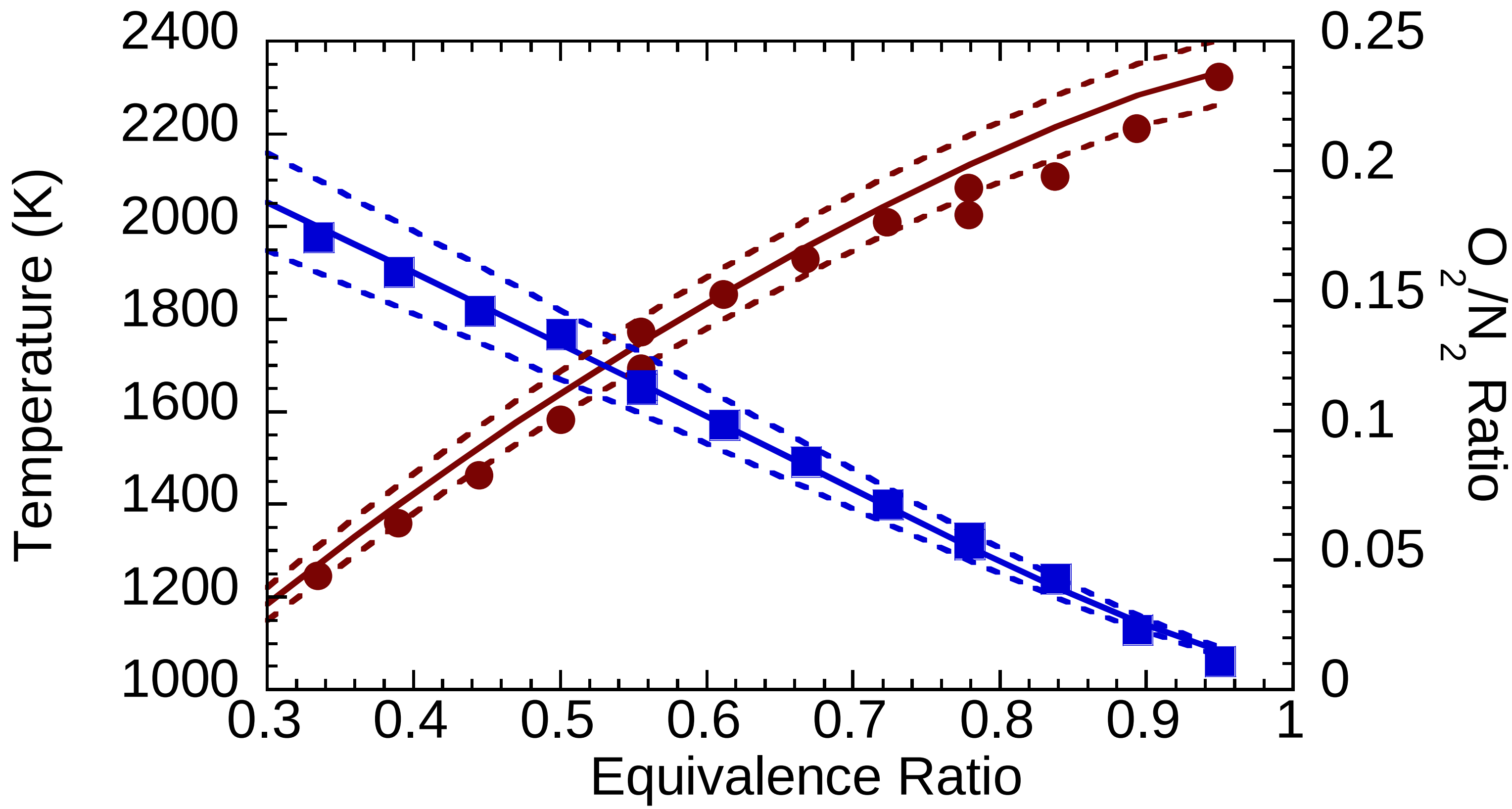


- N_2 contributions dominate all spectra
- O_2 sensitivity arises from alteration of spectral "envelope" and subtle line shifts

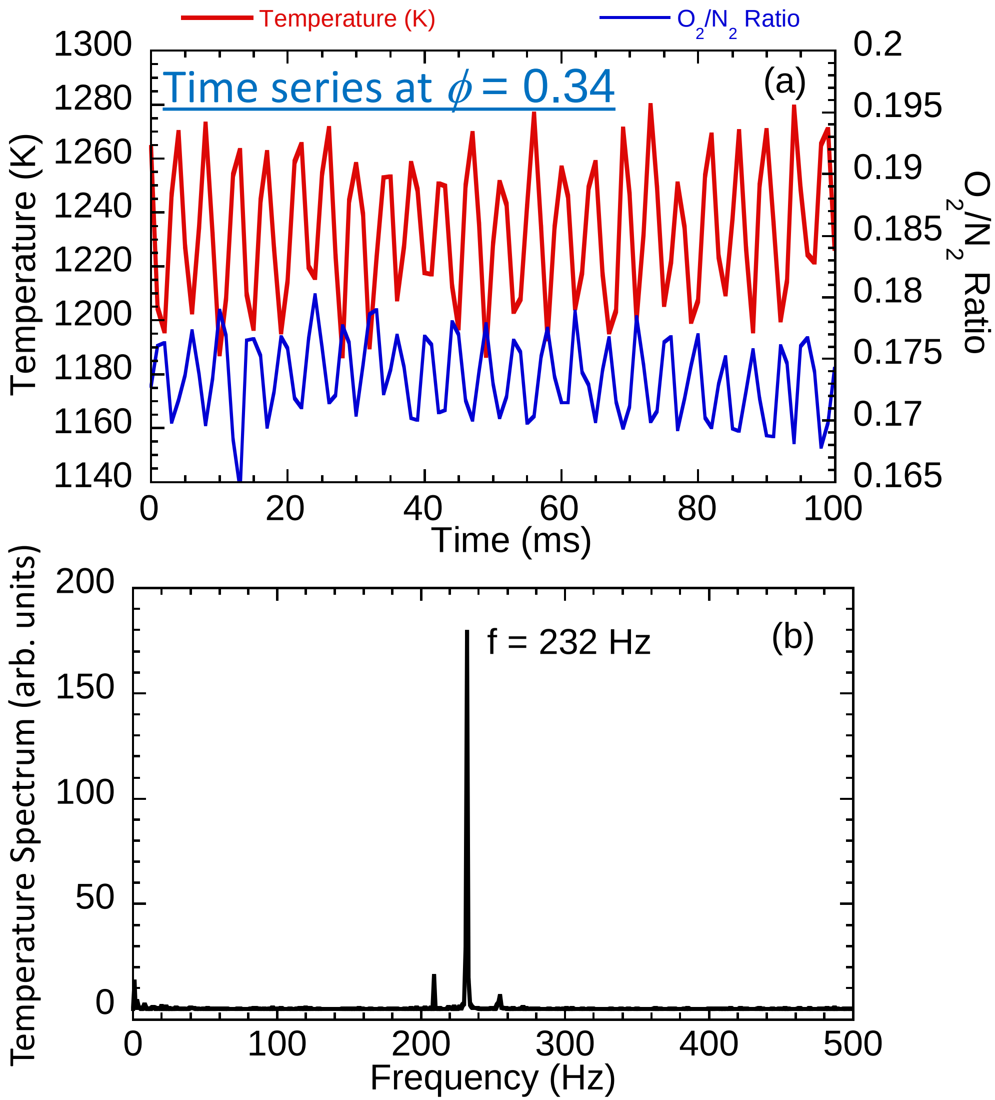
Kearney and Scoglietti, *Optics Letters* **38** (2013).

H_2/Air Flame Measurement Accuracy



- Temperature accuracy: ±3% when shot-averaged spectra are used
- Temperature accuracy: +3 to -6% when single-shot means are used
- O_2/N_2 accuracy is ±6% for ϕ up to ~0.65
- Uncertainty due to metered gas flows: Temperature ±3% / O_2/N_2 ±6-12%

kHz-rate dynamics in lean H2/air flames



- 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!*

C_2H_4/Air Flames on McKenna Burner

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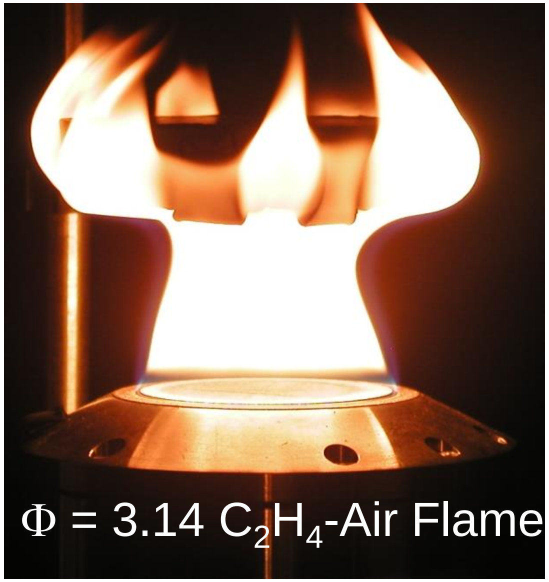
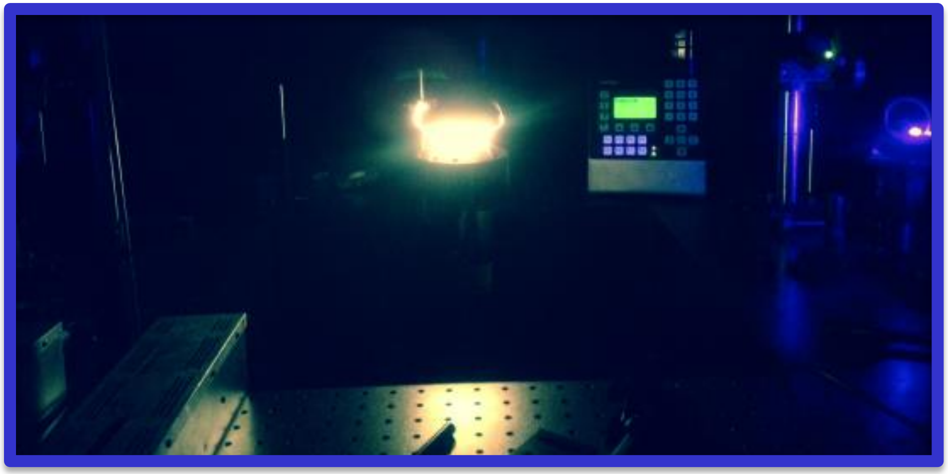
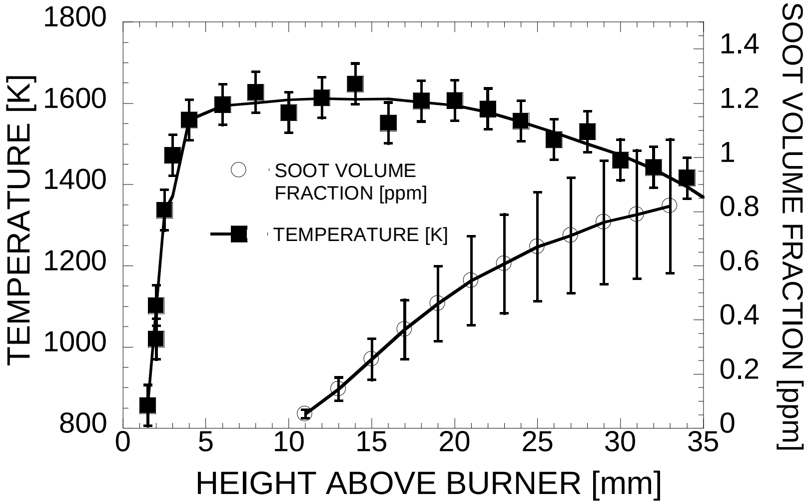
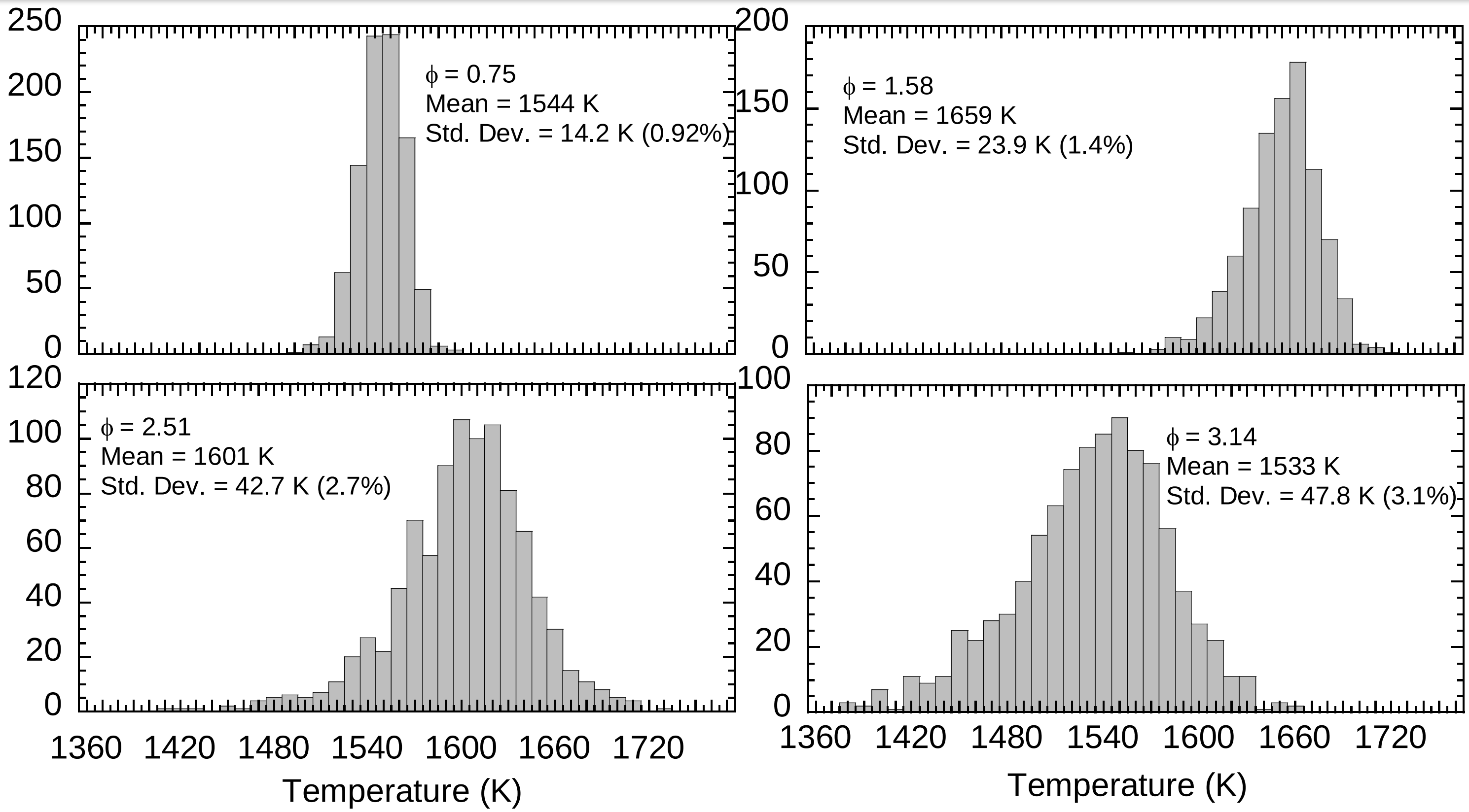
- Premixed hydrocarbon/air flame
- Water-cooled non-adiabatic burner
- Stable region ~5-15 mm above burner
- Previously studied at $\phi = 3.14$ in our lab (and elsewhere!)
- Wide range of stoichiometry, $\phi = 0.75$ to 3.14
- Potential contributions from N_2, O_2, CO, (CO_2 minimized by probe delay)







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

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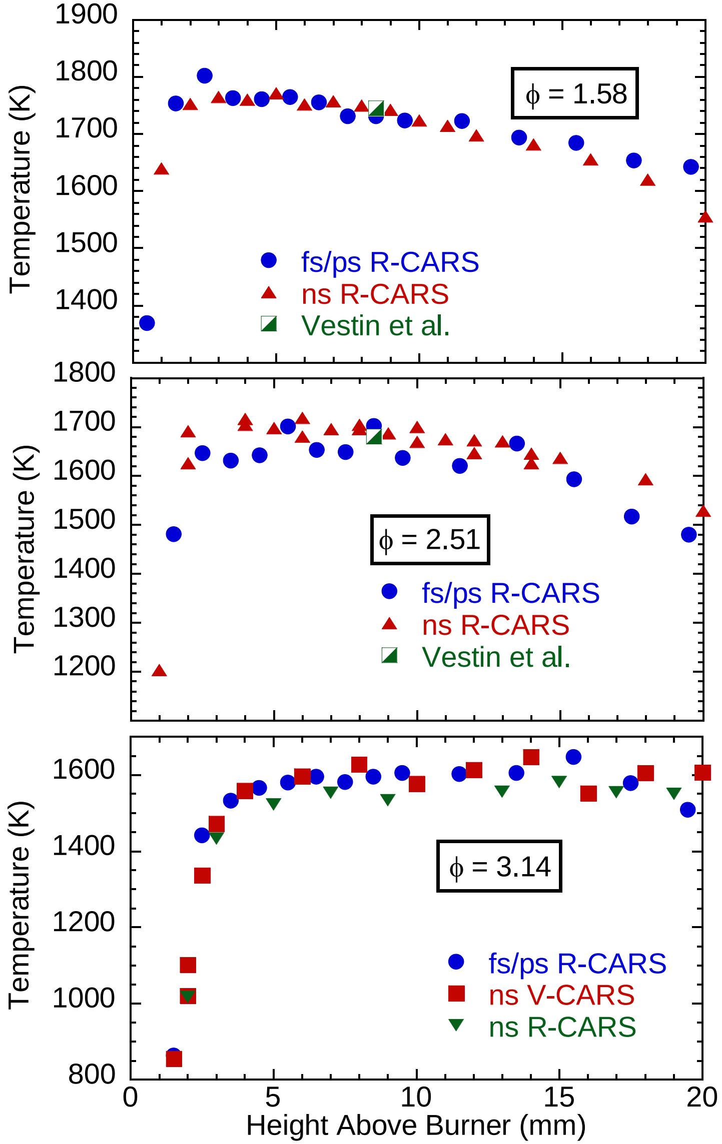
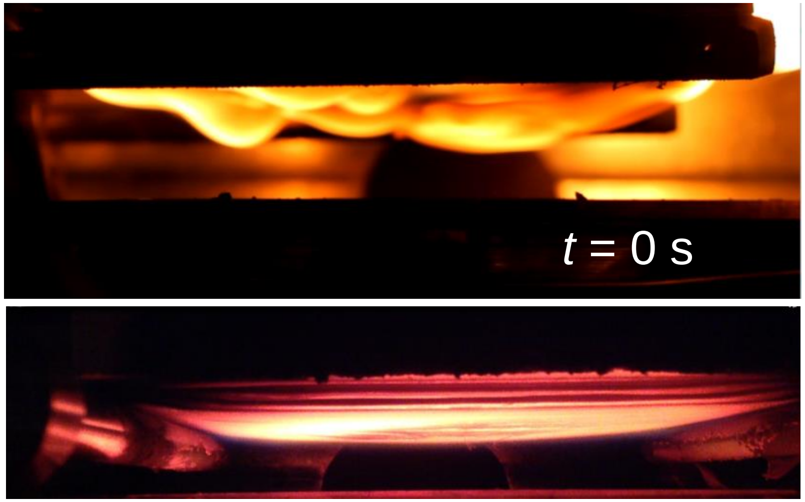


- Comparison is most valid in stable region 5-15 mm height
- ns-CARS temperatures based on fits to shot-averaged spectra
- Agreement of fs/ps and ns CARS is within 1-3% in these ***fuel-rich, sooting*** flames
- Within reported accuracy of rotational ns-CARS
- ns-CARS measurements
  - Rotational CARS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

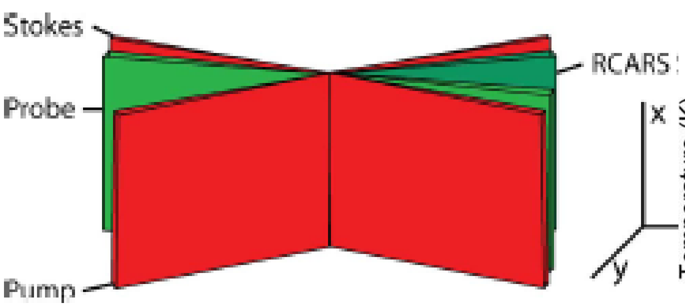
"Line CARS" Diagnostic at Sandia/CA

Sandia National Laboratories

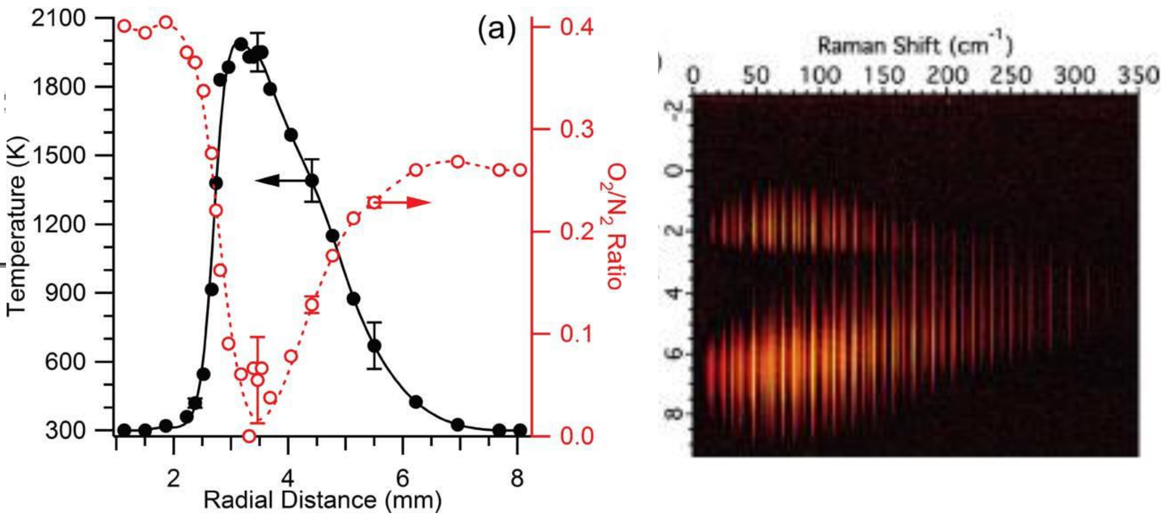
- Laser "sheets" form a linear beam crossing
- *The full spatial profile can be obtained on a single laser shot*
- *Temperature/O2 profiles obtained instantaneously!*
- 20 Hz data rate



Carbon-epoxy composite fire problem



CARS beam crossing



Temperature/O2 profiles from Propane/air flame

Kliewer et al, *Applied Optics* **50** (2011).

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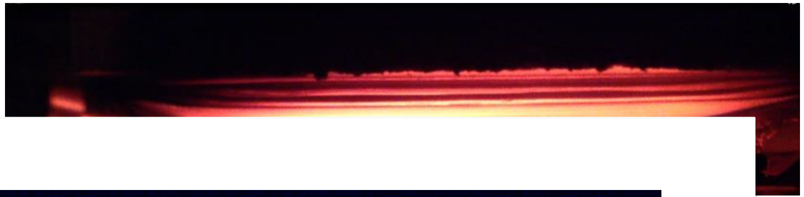

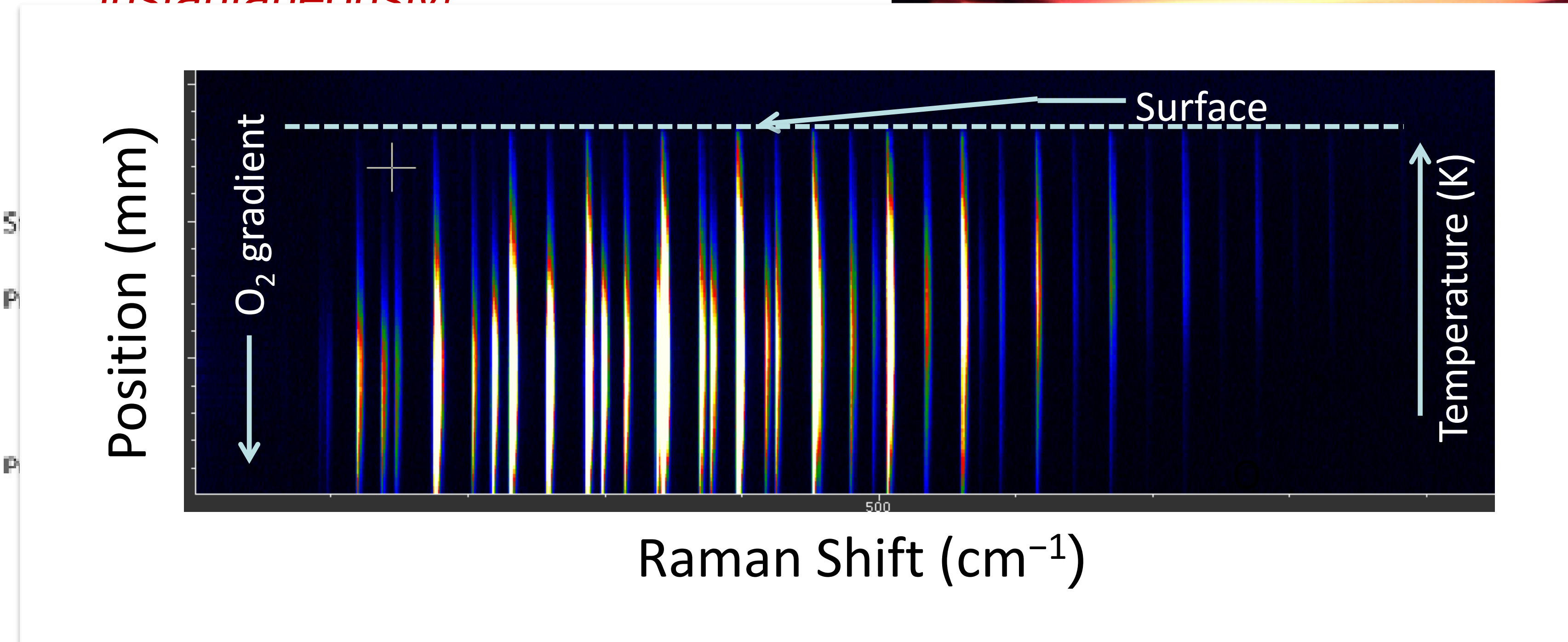

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Sandia National Laboratories

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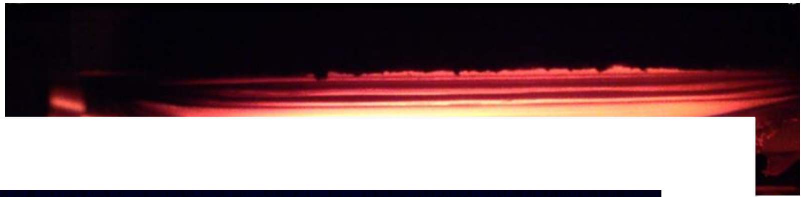
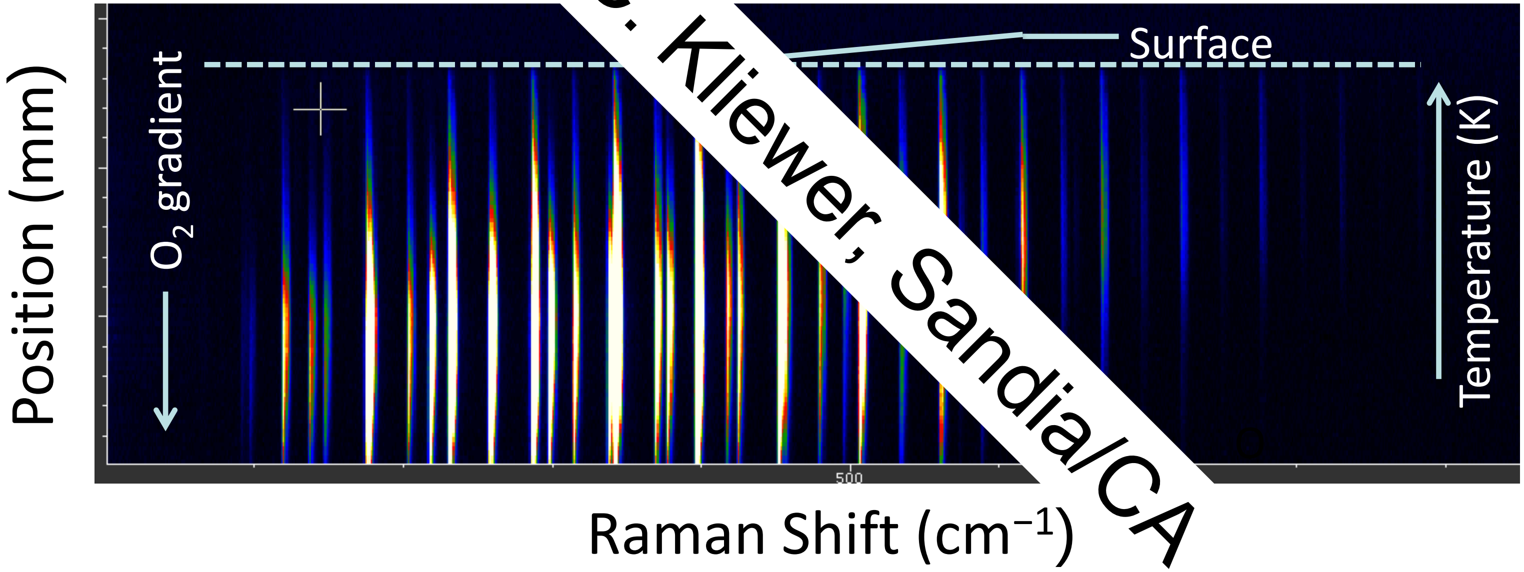




…em

300 350

Temperature/O2 profiles from Propane/air flame

Kliewer et al, *Applied Optics* **50** (2011).

Summary and Conclusions

- Femtosecond lasers are overcoming many traditional limitations in combustion diagnostics
  - "10-Hz barrier"
  - Collision-free measurements
  - High-quality broadband sources significantly improve precision
- We have performed a systematic assessment of fs/ps rotational CARS for kHz rate temperature/species data
- Temperature measurement precision is as good as 1%!
- Precision of O_2/N_2 ratio is 1-10% and monotonically increases with O_2/N_2
- We have demonstrated SHBC for high-energy kHz rate probes
  - Enabling technology for high-rate temperature/species measurements
  - Flame dynamics observed at ~250 Hz
  - Successful demonstration in sooting flames
- Collaboration with SNL/CA for line-imaging measurements

Selected Publications and Presentations

Femtosecond rotational CARS development at kHz rates

Kearney, S.P., D.J. Scoglietti, and C.J. Kliewer, "Hybrid fs/ps Rotational CARS Temperature and Concentration Measurements Using Two Different ps-Duration Probe Beams," *Optics Express* **21**, 12327-12339, 2013.

Kearney, S.P. and D.J. Scoglietti, "Hybrid fs/ps Rotational Coherent Anti-Stokes Raman Scattering at Flame Temperatures Using a Second-Harmonic Bandwidth-Compressed Probe, *Optics Letters* **38**(6), 833-835, 2013.

Kearney, S.P. and D.J. Scoglietti, "Accuracy and Precision of kHz-rate Rotational fs/ps CARS in the products of hydrogen and ethylene air flames," *submitted to Combustion and Flame,* 2014.

Large-scale hydrocarbon fire measurements

Kearney, S.P., K. Frederickson, and T.W. Grasser, "Dual-Pump Coherent Anti-Stokes Raman Scattering Thermometry in a Sooting Turbulent Pool Fire, *Proceedings of the Combustion Institute* **32**, 871-878, 2009.

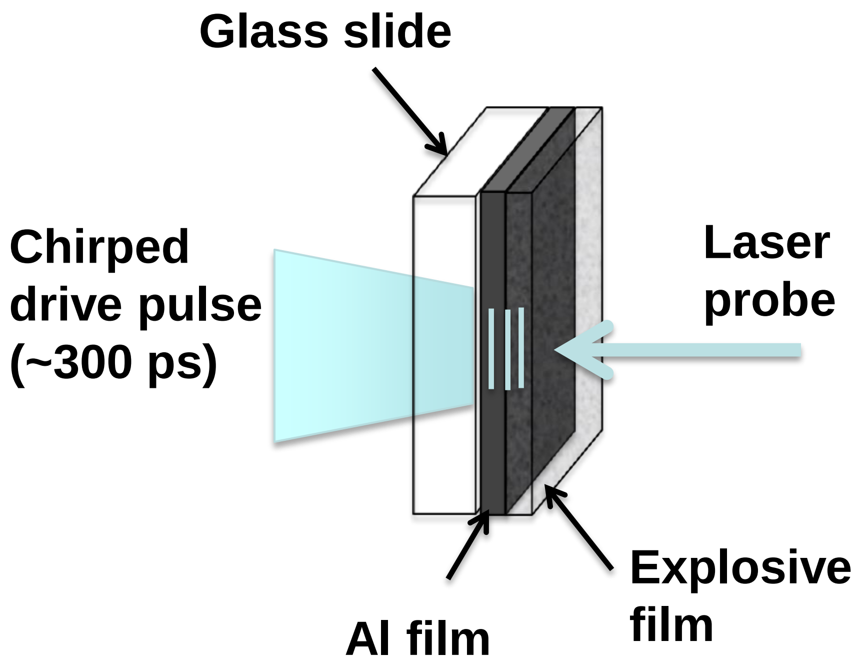
Frederickson, K., S.P. Kearney, A. Luketa, J.C. Hewson, and T.W. Grasser, "Dual-Pump CARS Measurements of Temperature and Oxygen in a Turbulent Methanol-Fueled Pool Fire," *Combustion Science and Technology* **182**, 941-959, 2010.

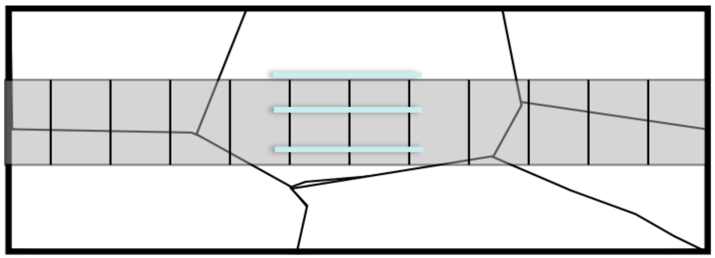
Kearney, S.P. and F. Pierce, "Evidence of Soot Superaggregates in a Turbulent Pool Fire," *Combustion and Flame* **159**, 3191-3198, 2012.

3 Invited Talks (USU/Uva/LACSEA) in FY2014

Laser Drive Experiments

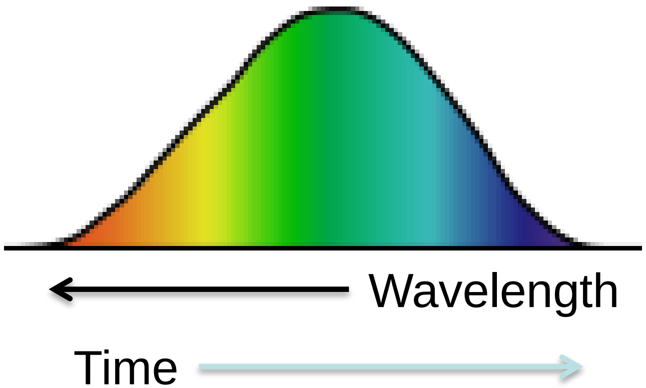
- Ultrafast laser shock drive experiments on thin film explosives.
- Image mechanical response (USI).
- Image thermochemical response (SRS/Transient Absorption).

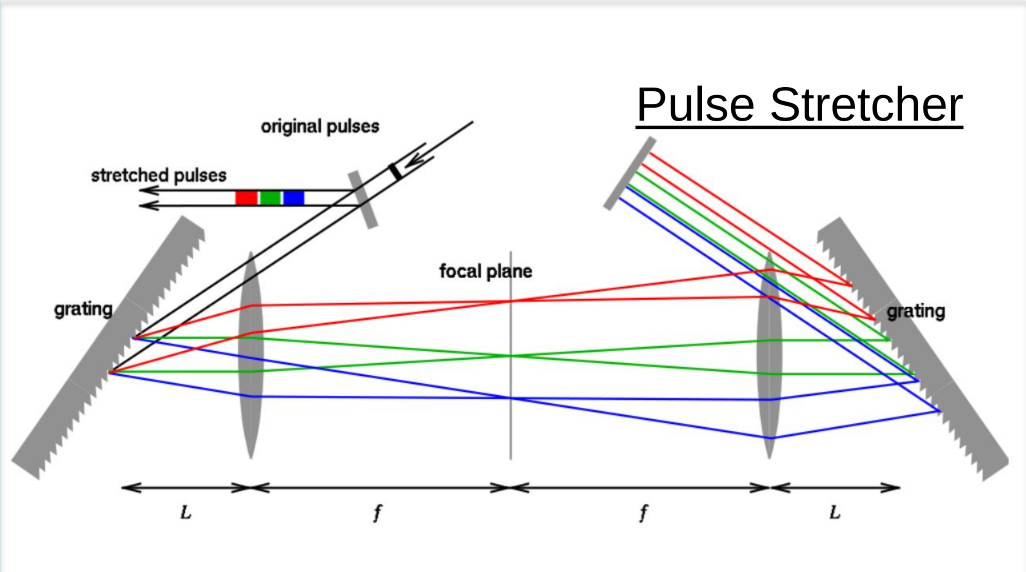


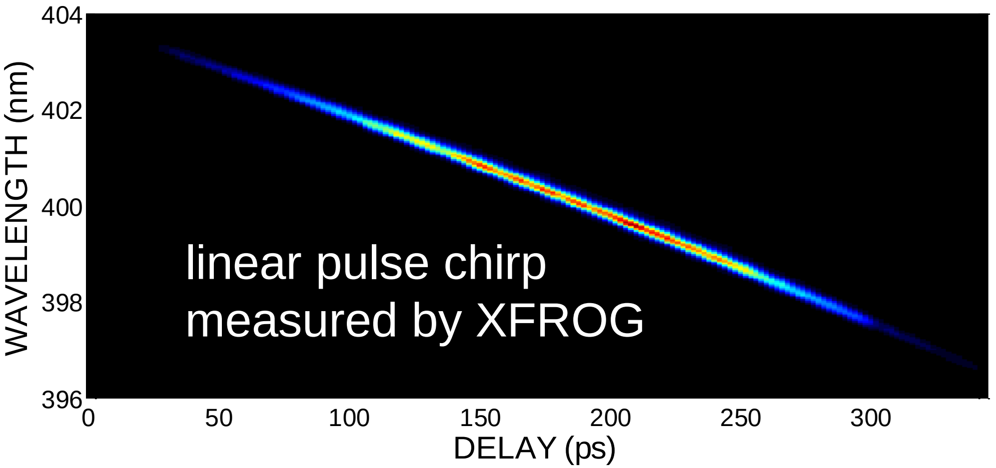


Femtosecond laser pulses have wide frequency spectra that allow us to encode time into frequency (color)

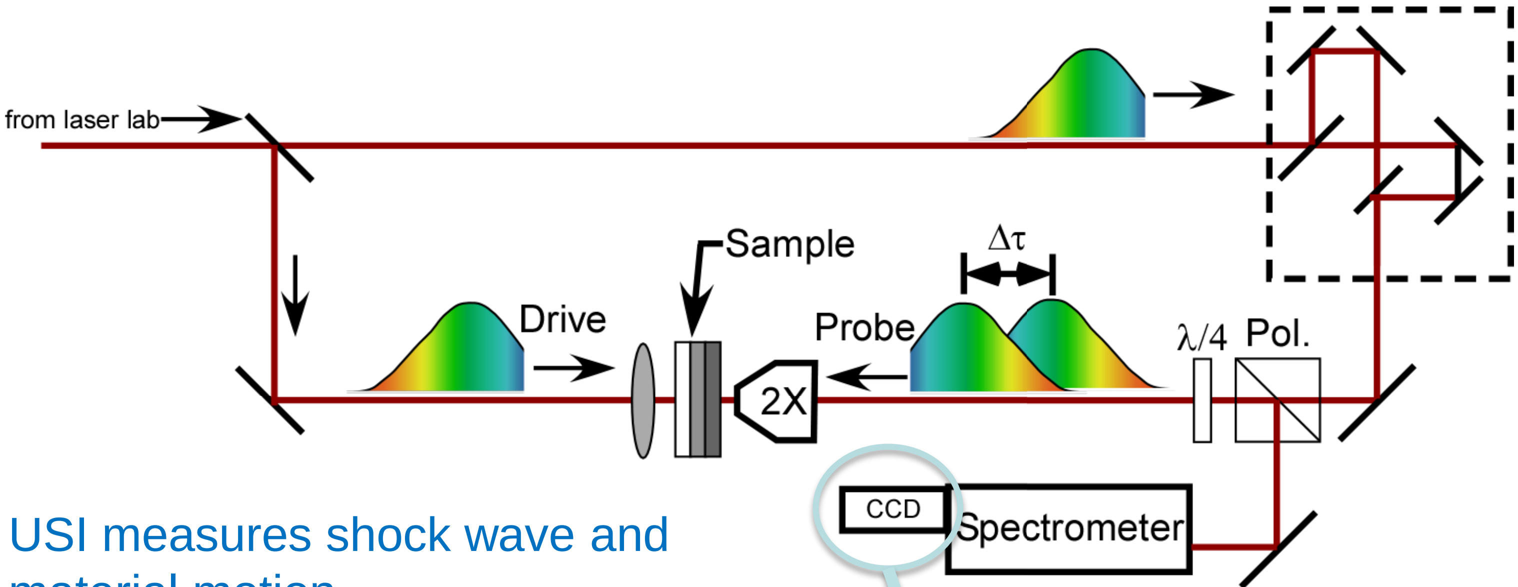
- Femtosecond bandwidths are typically 10s of nanometers
- Can be readily compressed or stretched (chirped) using grating-based stretchers
- Linear chirp is added in our lab to **encode wavelength into time with picosecond resolution**



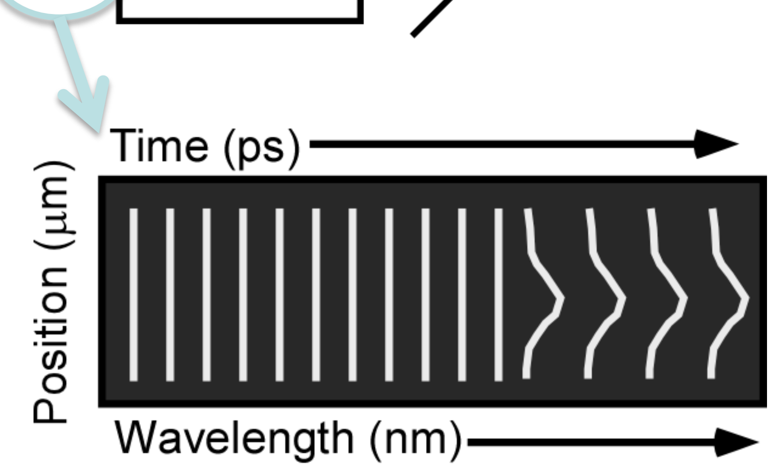




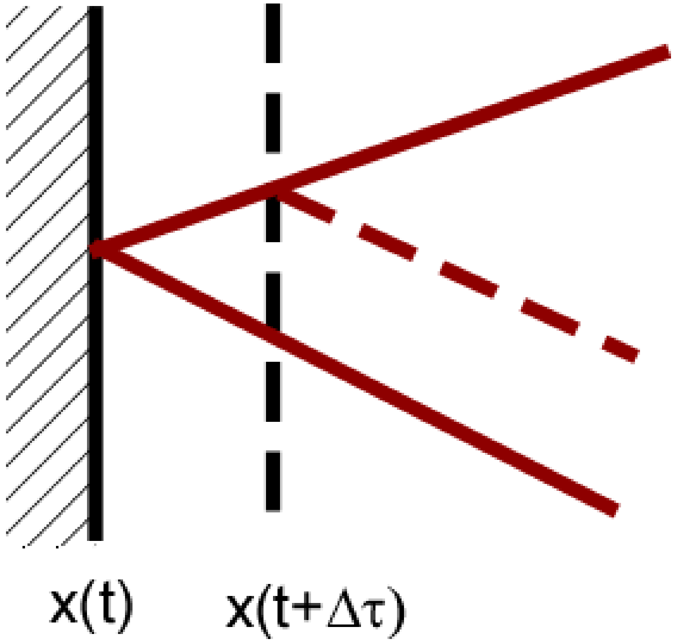
Ultrafast Shock Interferometry (USI) for characterization of shockwave structure and EOS measurements

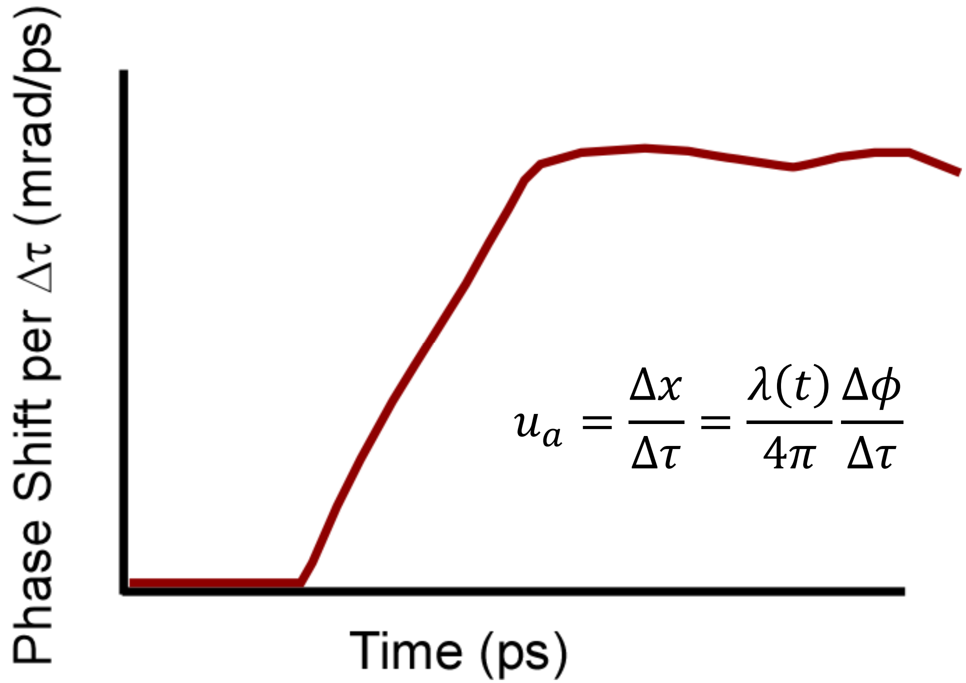


- USI measures shock wave and material motion
- Picosecond time resolution
- Micron-scale spatial resolution
- Laser-driven shock supported for 300-400 ps
- Shock-induced phase shift accumulated during probe separation, $\Delta\tau$, is monitored



Canonical USI response on bare metal ablator

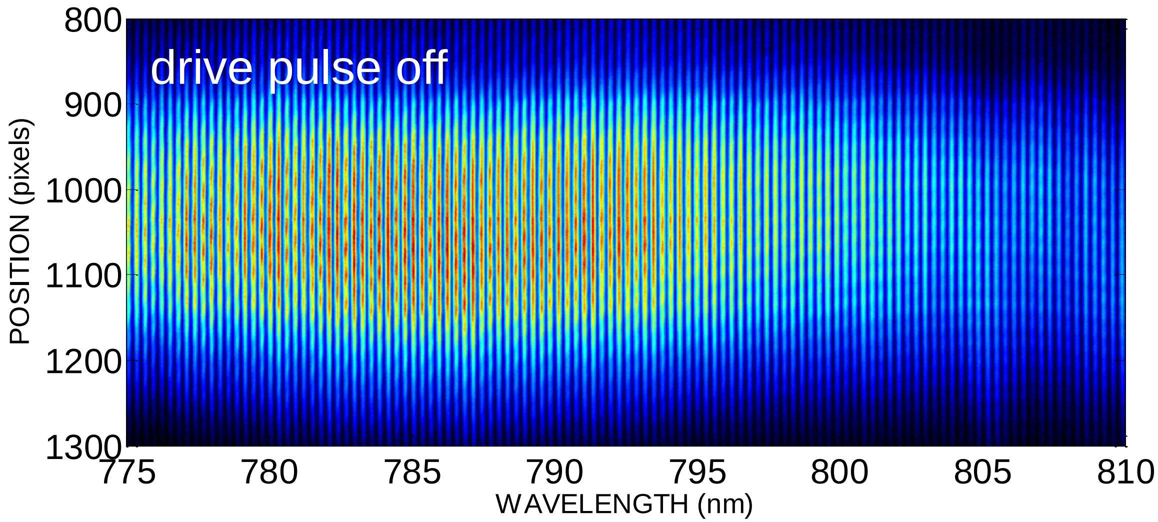


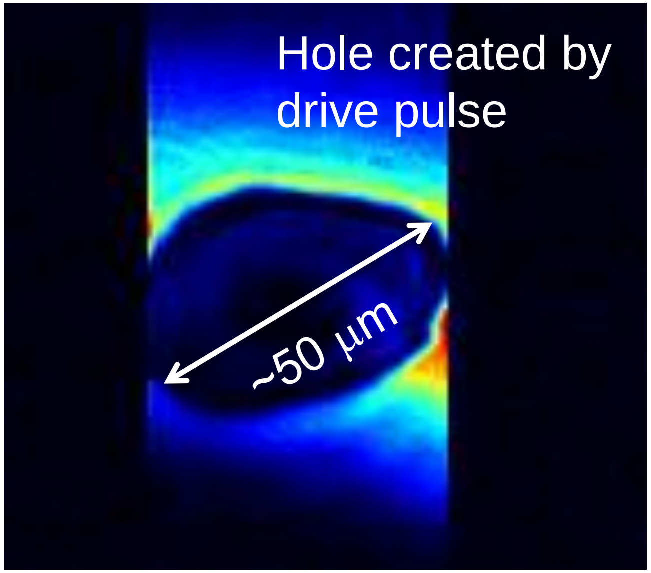


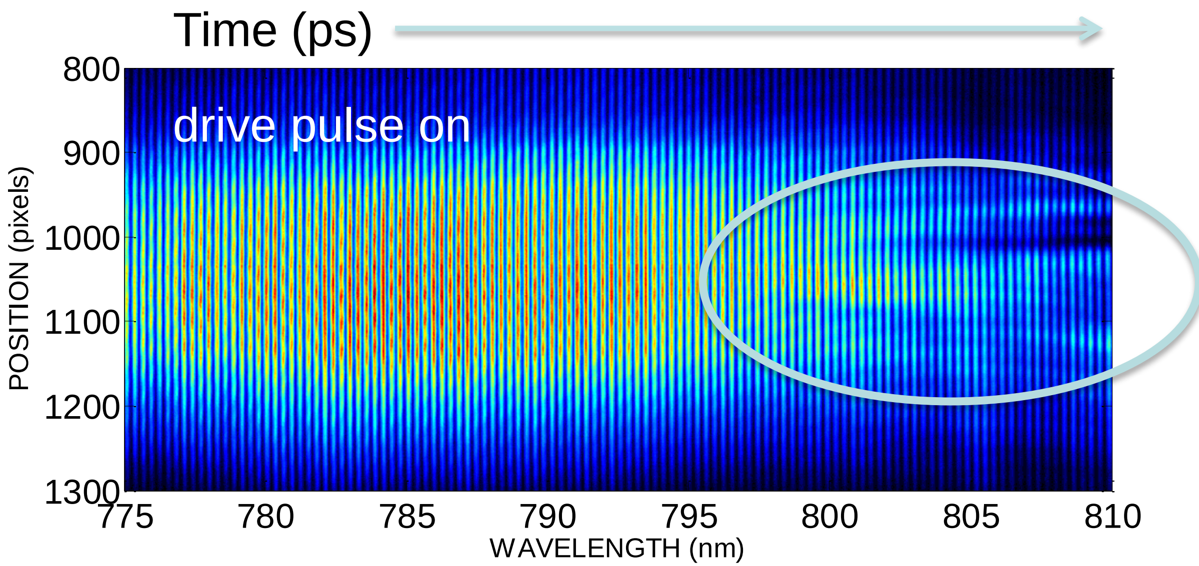
$$u_a = \frac{\Delta x}{\Delta \tau} = \frac{\lambda(t)}{4\pi} \frac{\Delta \phi}{\Delta \tau}$$

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