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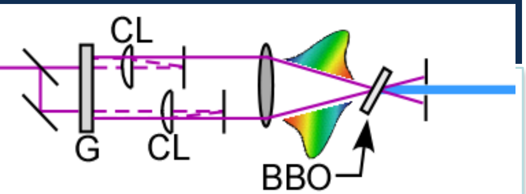

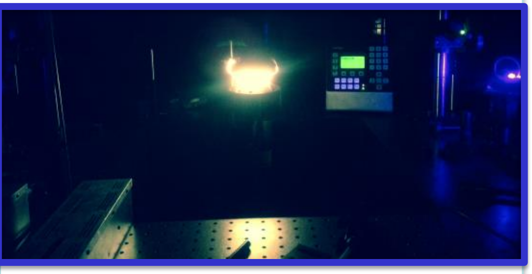

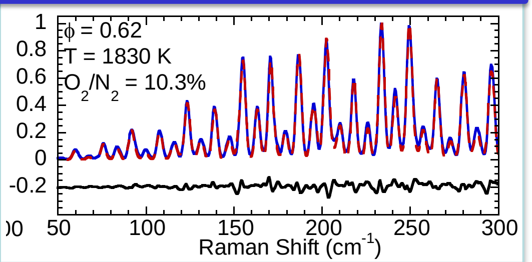

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Exceptional service in the national interest

Hybrid fs/ps Rotational CARS Temperature/Species Detection in Flames at kHz Rate

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U.S. DEPARTMENT OF ENERGY

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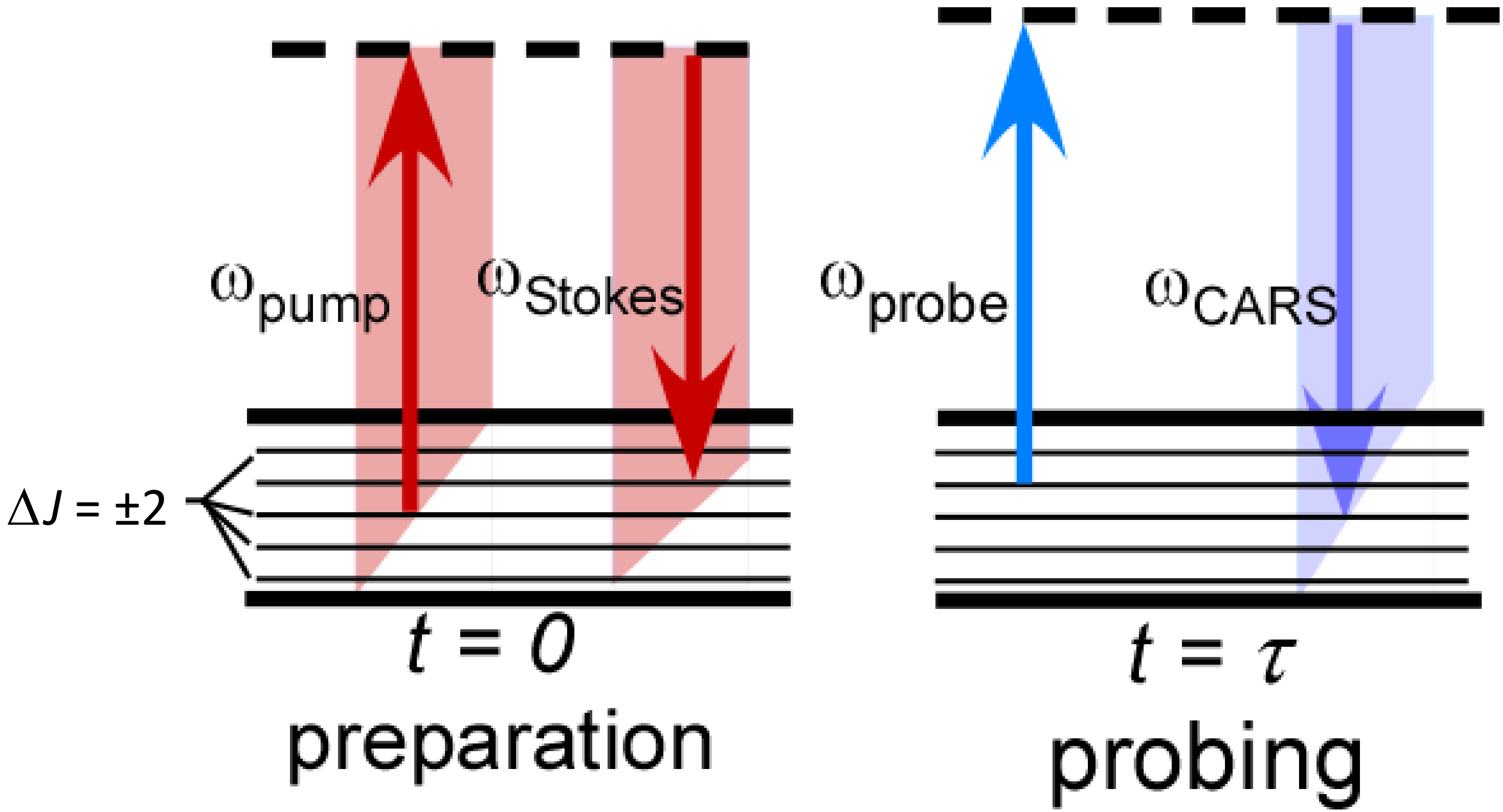
Motivation and Outline of Presentation

Motivation: Verify the <u>accuracy and precision</u> of fs/ps rotational CARS for

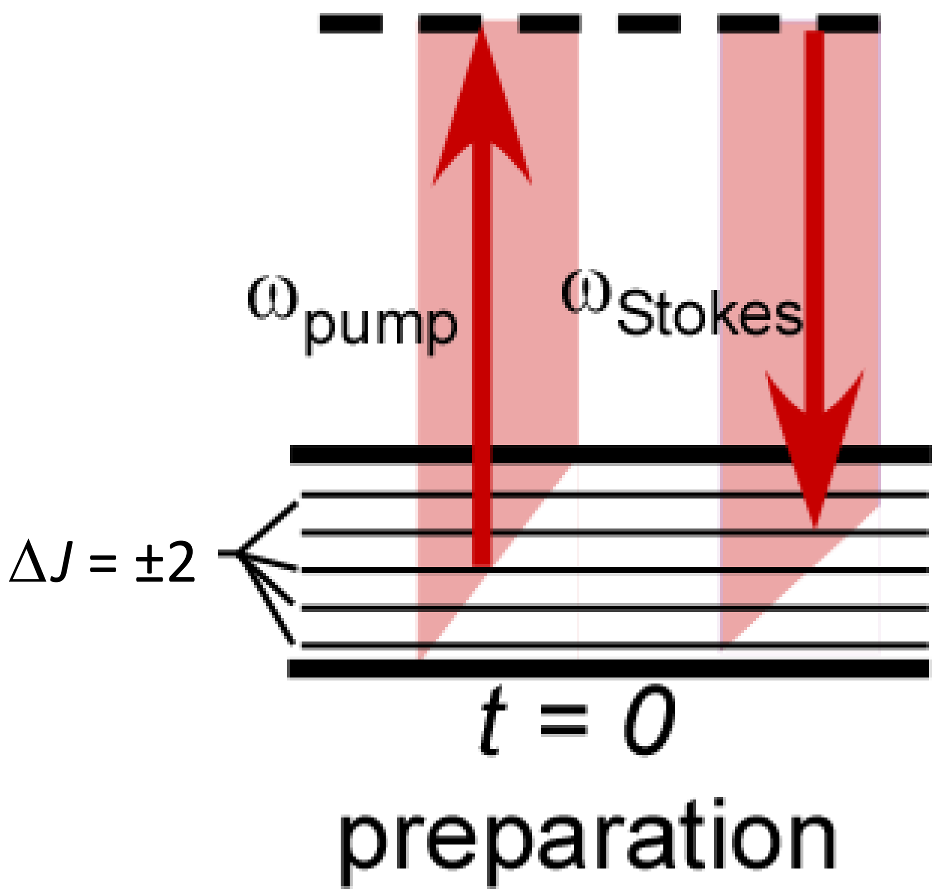
- Temperature *<u>and</u>* O_2 concentration
- A wide range of stoichiometry
- Hydrogen and hydrocarbon fuels
- Sooting conditions
- Single-laser-shot measurements at 1 kHz

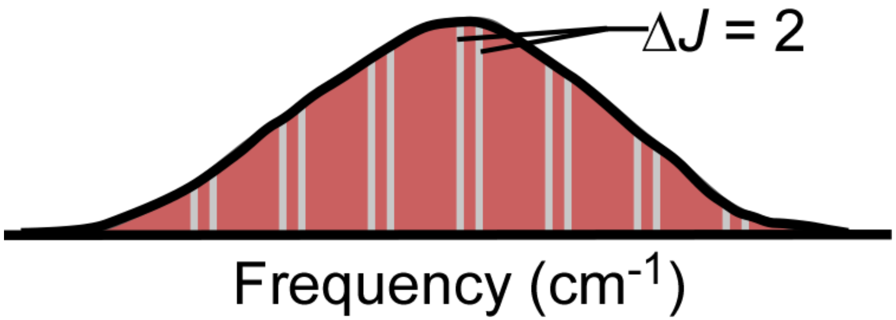
- <u>fs CARS processes…2 key points!</u>
  - *<u>Two-photon Raman preparation</u>* at time $t = 0 \rightarrow$ high-precision
  - High-energy picosecond probing by *<u>second-harmonic bandwidth compression (SHBC)</u>*
- <u>Experiments to assess accuracy and precision</u>
  - H_2/air flat flames – compare to equilibrium
  - C_2H_4/air flat flames – compare to ns CARS
- <u>Summary and Conclusions</u>
- <u>Bonus material: Pressure Measurements</u>

Time-Domain Rotational Raman



fs pulses provide low-noise preparation

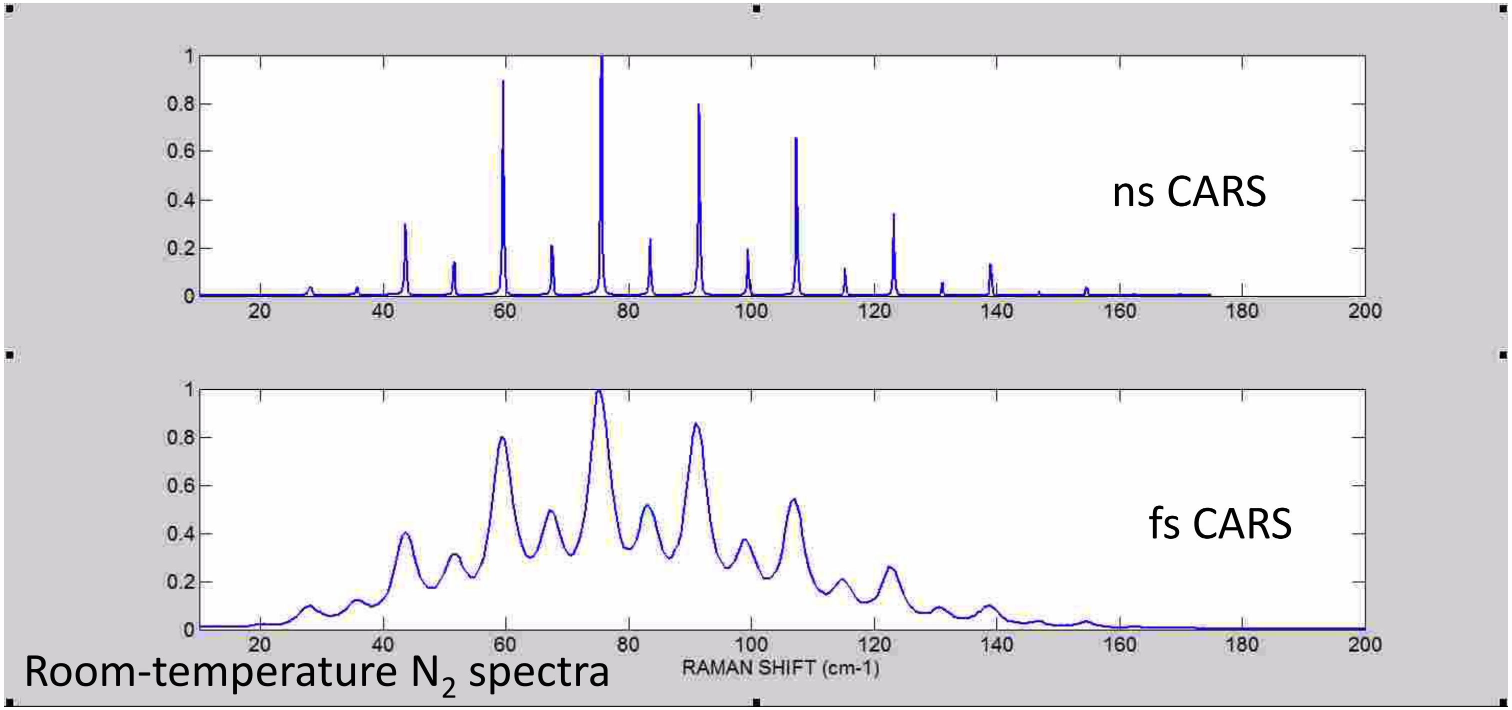




"Two Photon Effect"

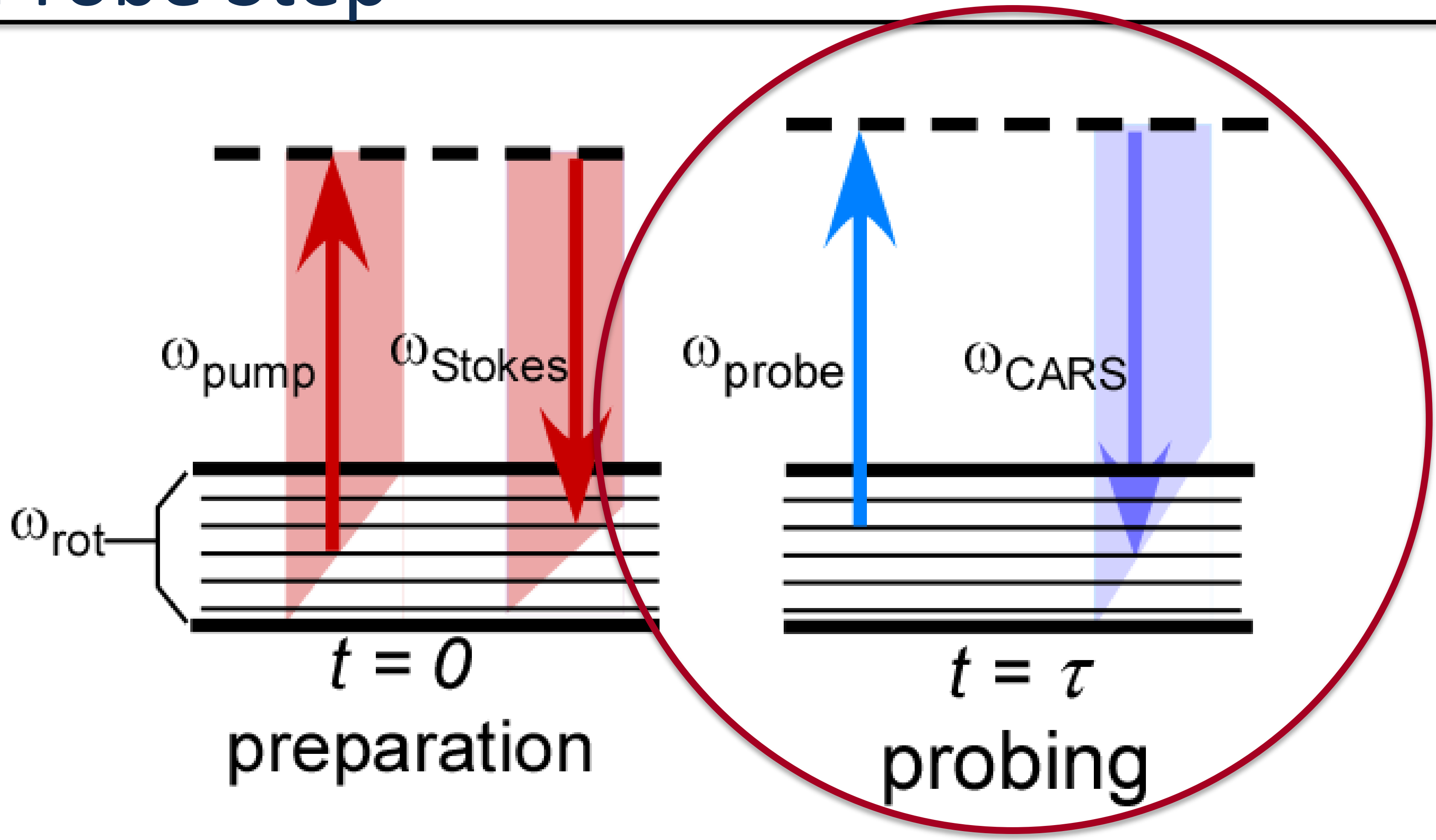
- Large number of frequency combinations prepare each Raman transition
- Fourier-transform limited pulses
  - Flat spectral phase
  - Low phase and amplitude noise
  - Low noise/high efficiency

Can rotational fs/ps CARS be applied in flames?



- Precision in flame temperatures approaching 1% using <u>vibrational</u> CARS
  - Richardson *et al.*, Roy *et al.*
- Similarly outstanding precision reported for <u>rotational</u> CARS ***<u>below 800 K</u>***
  - Stauffer *et al.*, Kearney *et al.*
- **Can rotational Raman be used at flame temperatures?**
- **Temperature *<u>and concentration</u>* at kHz rates?**

Probe Step

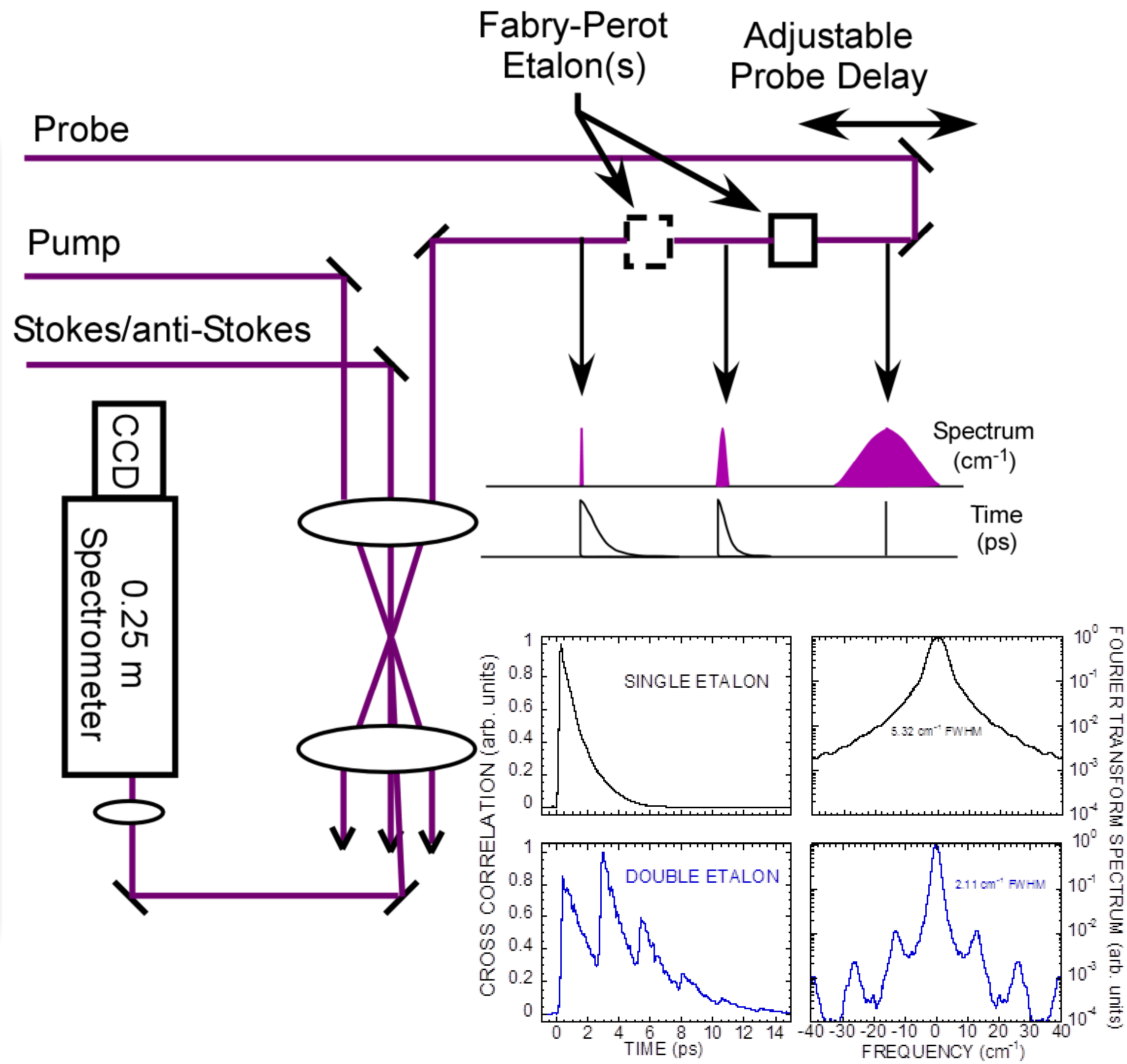


Probe Pulse Requirements:

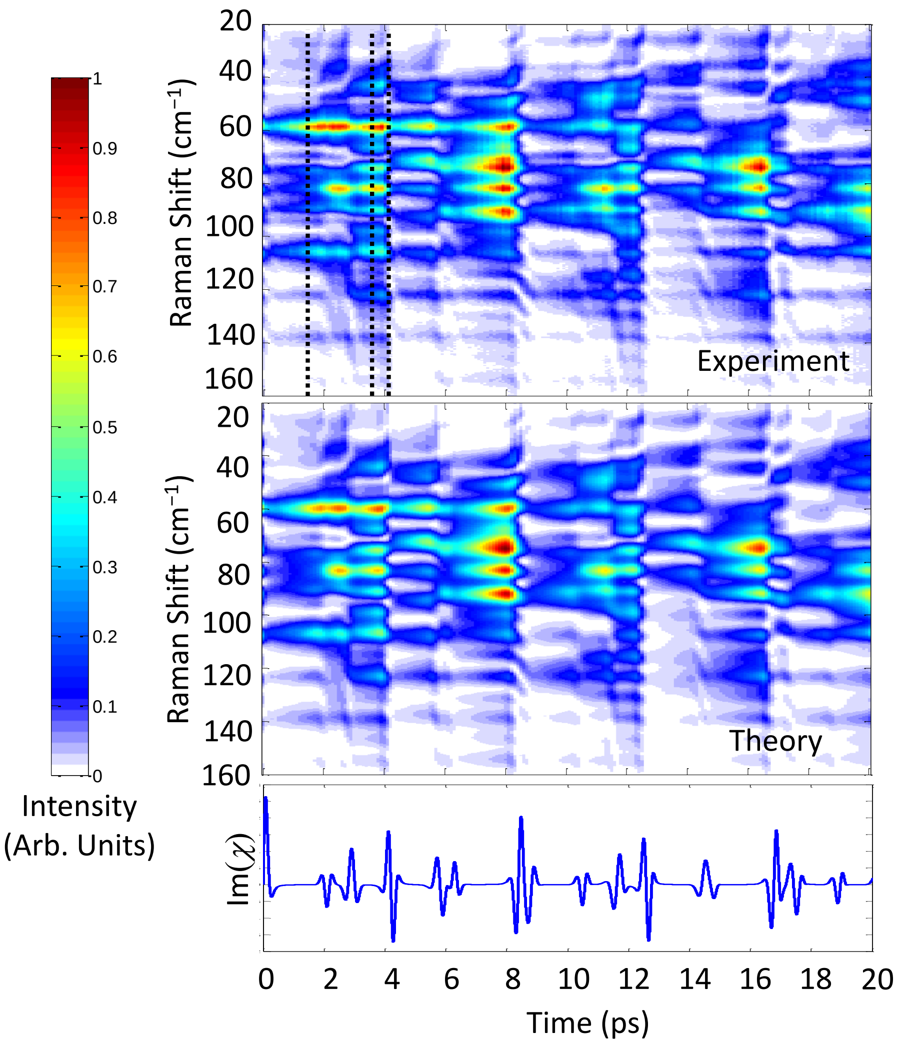
- ***Phase locked*** to fs preparation pulses
- ***Frequency narrow*** to resolve Raman spectral features

"Bandwidth Carving" Experiments

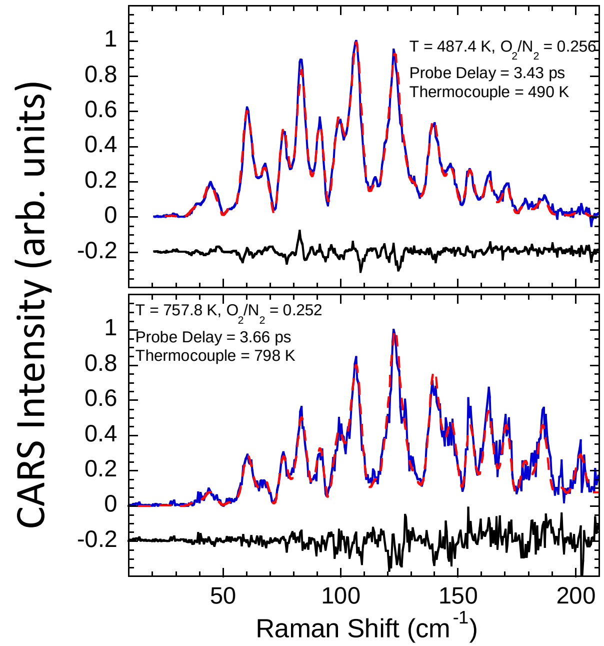
- **"Bandwidth-Carving"** to generate ps probe pulse
- Two different probe resolutions investigated with single- and double-etalon configurations
- Very inefficient (0.8 to 2.4% or less transmission)
- Atmospheric air spectra in tube-furnace up to 800 K for probe delays up to 20 ps



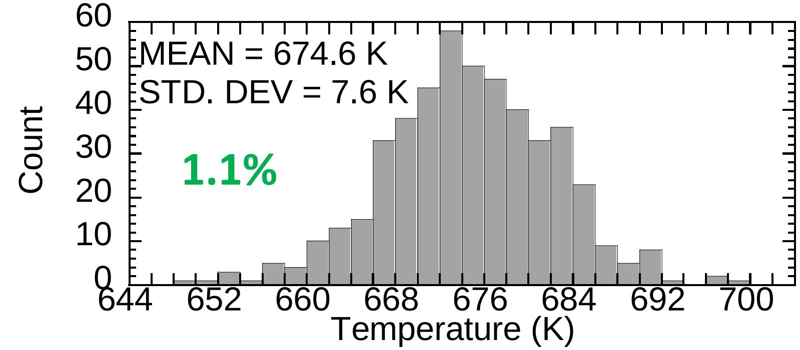
Results with etalon-generated ps probe beam



Room-temperature coherence dynamics



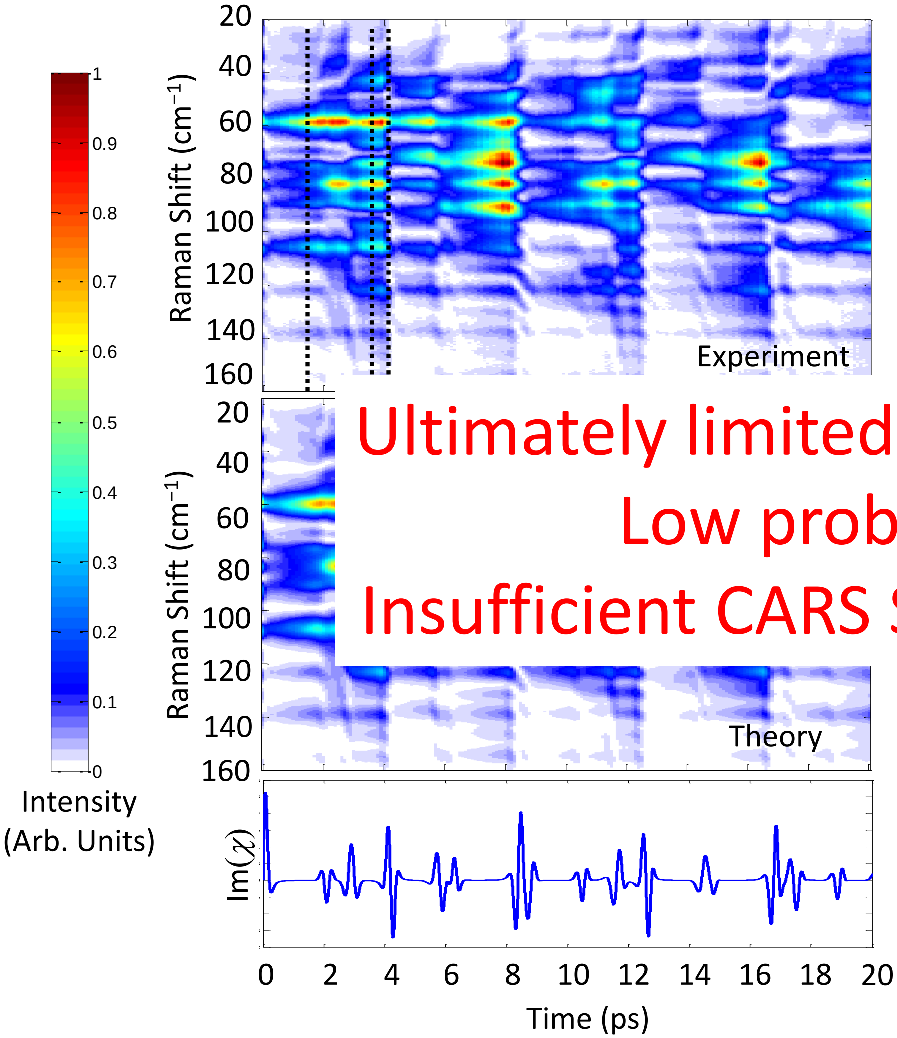
Spectra from tube-furnace-heated air



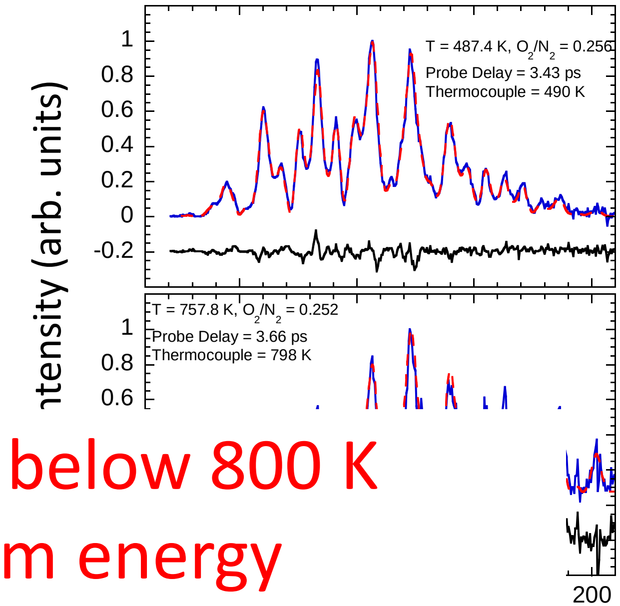
High-precision thermometry

Kearney *et al.*, *Opt. Express* **21** (2013)

Results with etalon-generated ps probe beam



Room-temperature coherence dynamics

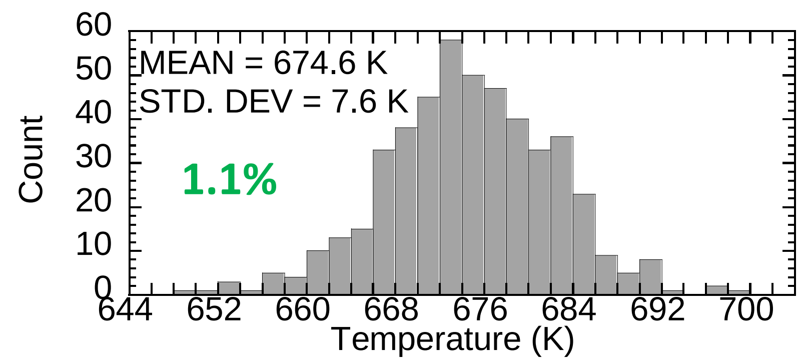


Ultimately limited to T below 800 K

Low probe-beam energy

Insufficient CARS SNR for single-shot data

ated air



High-precision thermometry

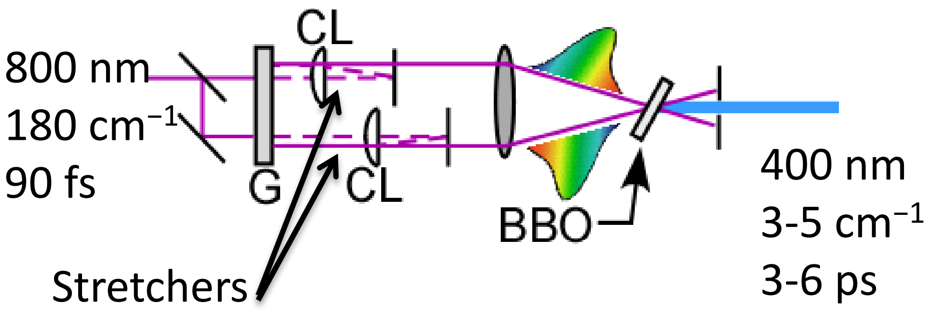
Second-Harmonic Bandwidth Compression (SHBC)

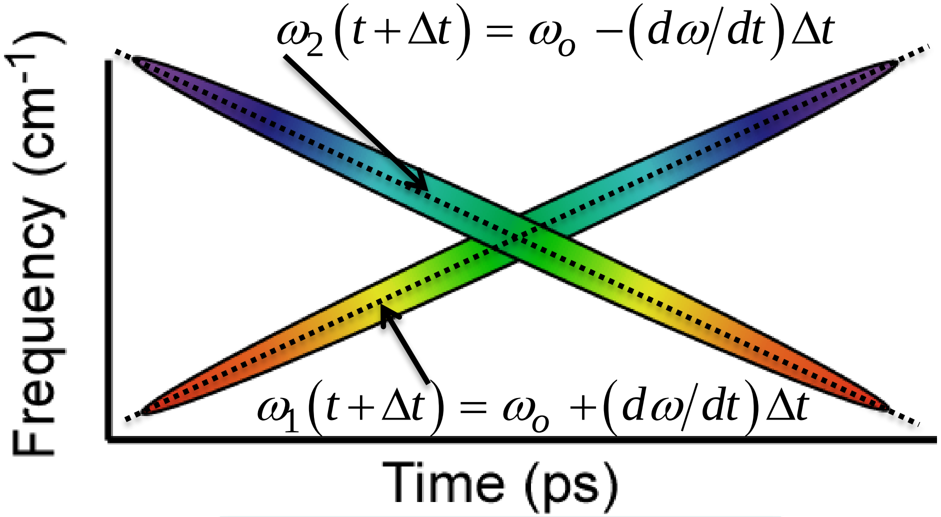
- Commercial device (Light Conversion)
- Converts fs radiation at 800 nm to ps radiation at 400 nm
- Grating pulse stretchers
- **<u>Phase-conjugate</u>** temporal chirps imparted upon broadband fs pumps
- Sum-frequency generation in BBO
- Output linewidth 3.5-4.0 cm^{-1}

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

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

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

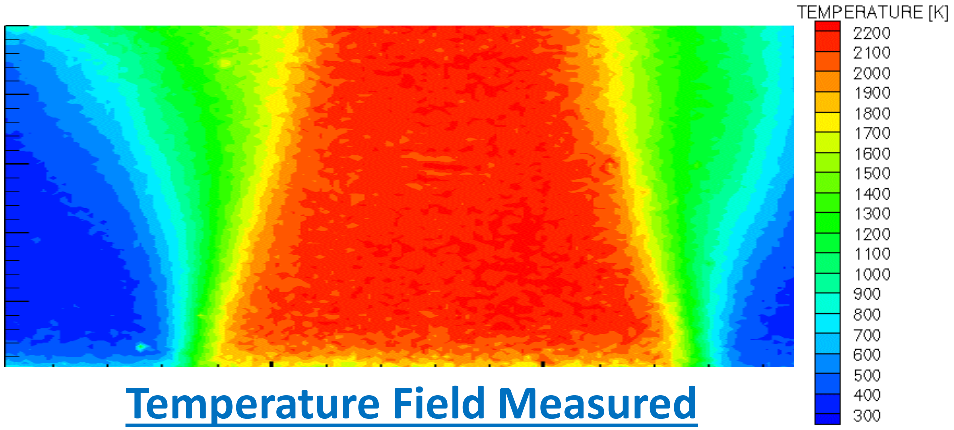




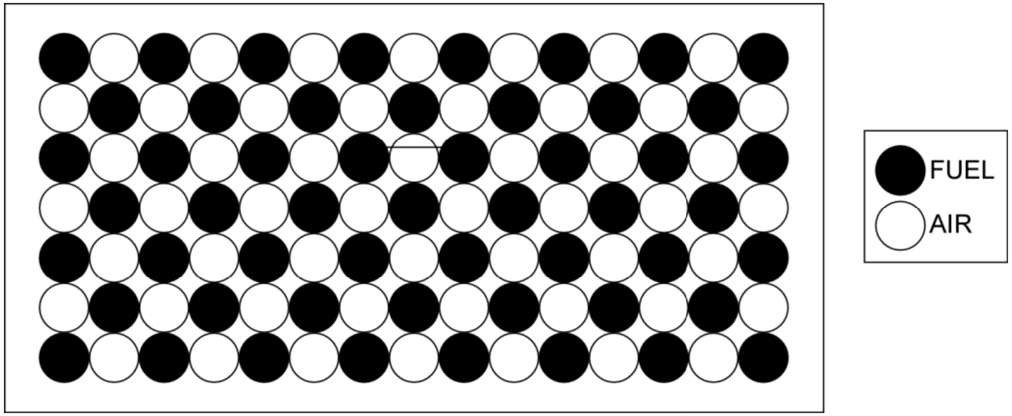
$$\boxed{\omega_{sfg} = \omega_1 + \omega_2 = 2\omega_o}$$

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 adiabatic equilibrium***

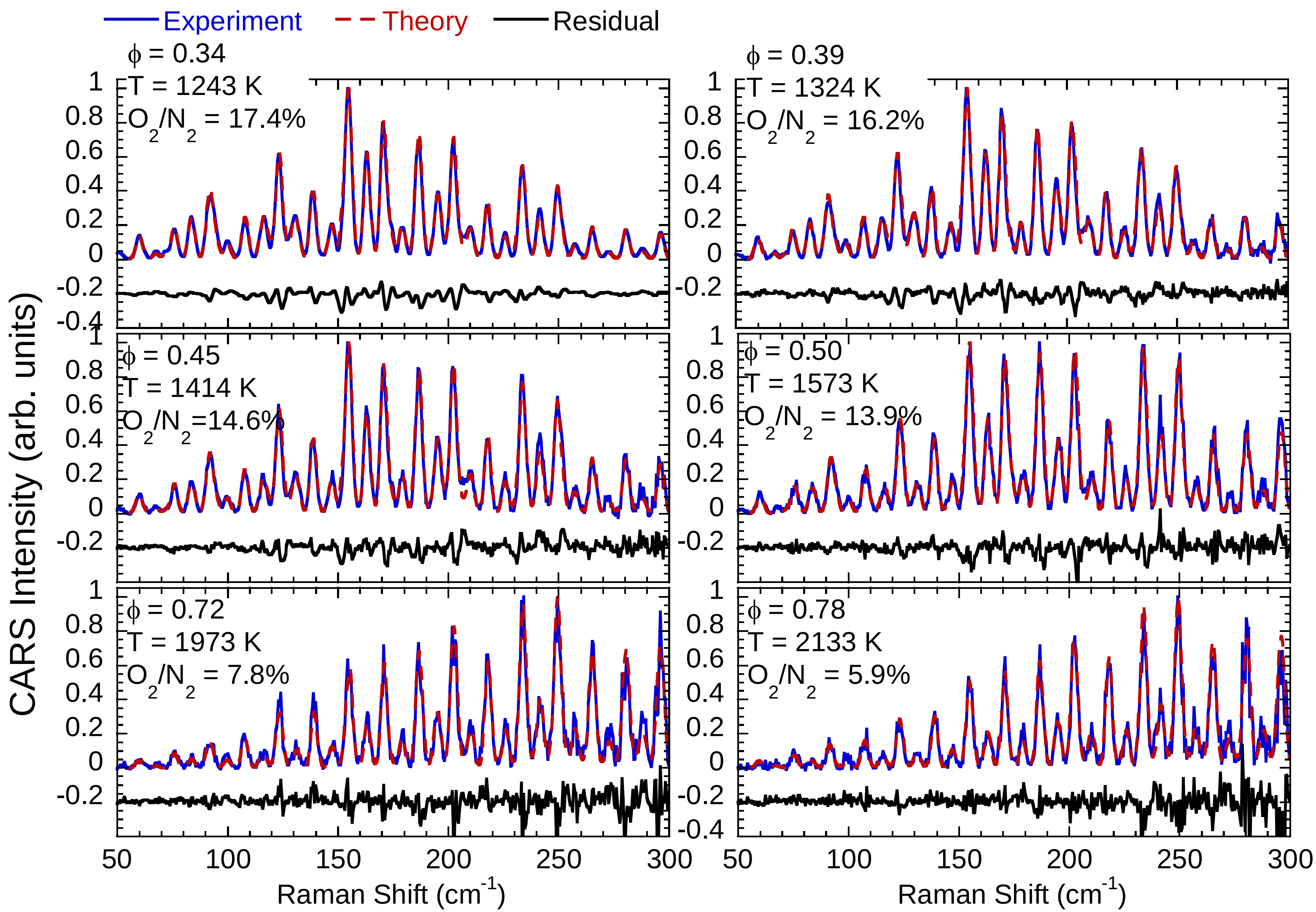


Temperature Field Measured by Rayleigh Scattering (CH_4/air)



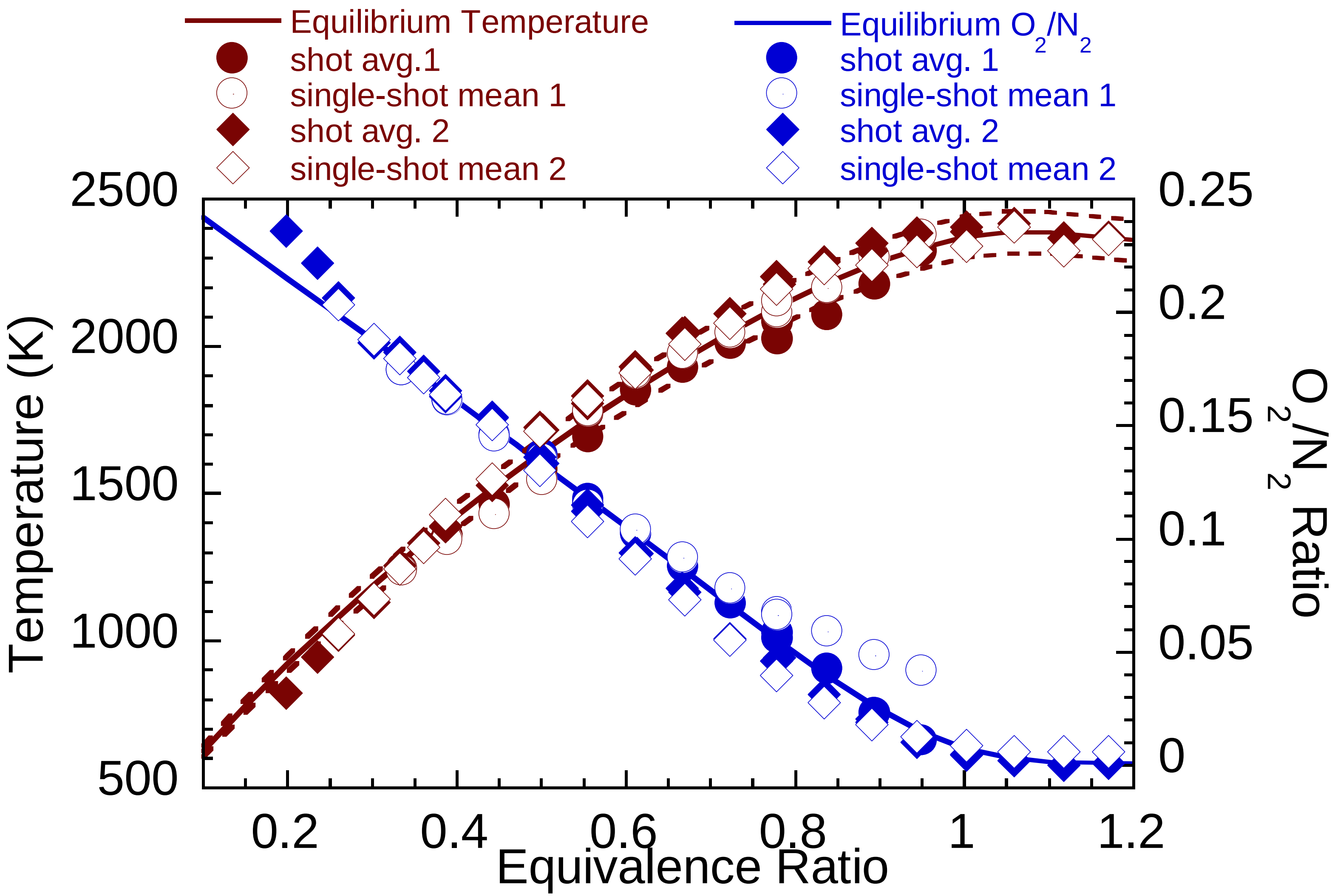
Burner Hole Configuration

Single-Shot Spectra from Near-Adiabatic H2/air flame

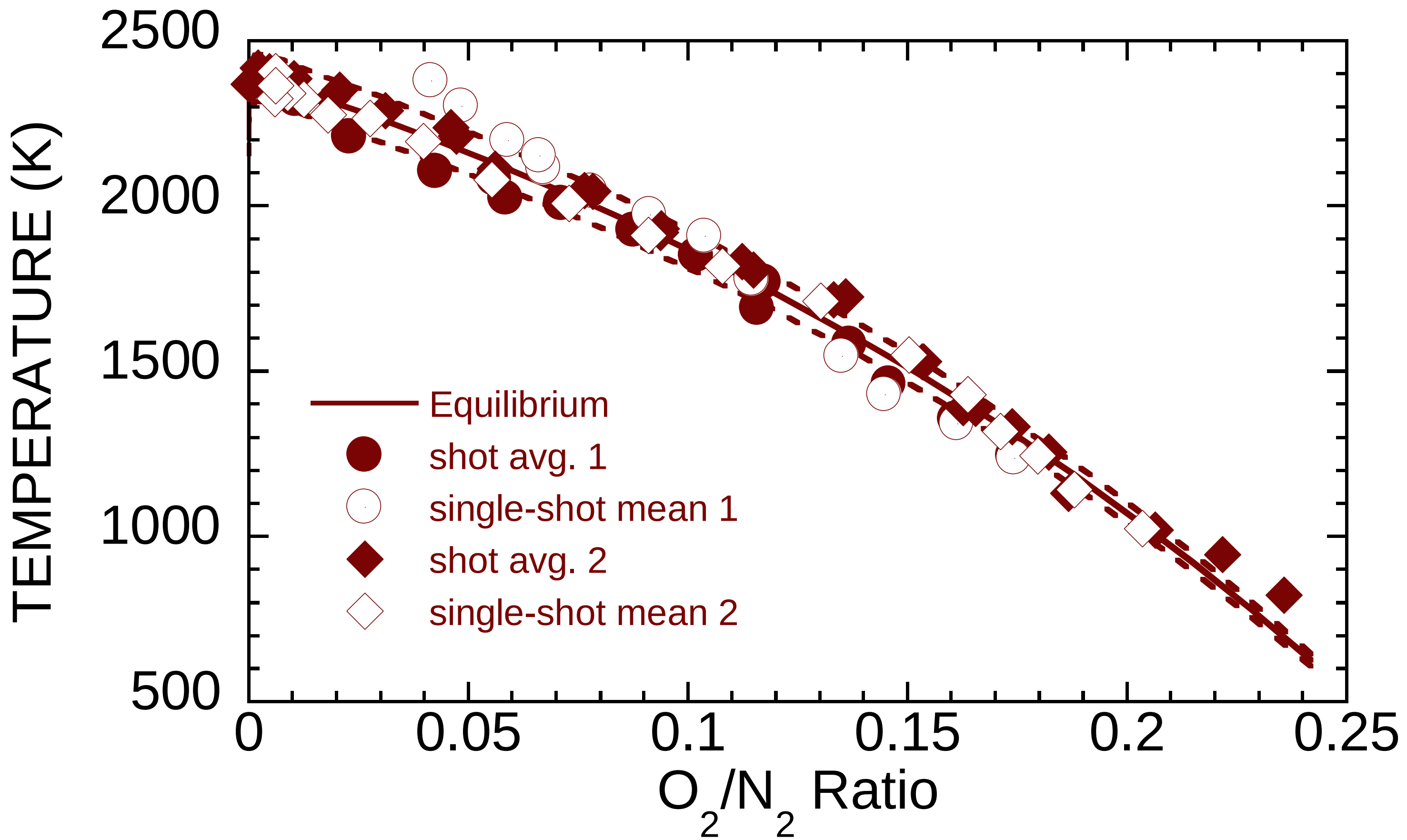


Kearney and Scoglietti, *Opt. Lett.* **38** (2013).

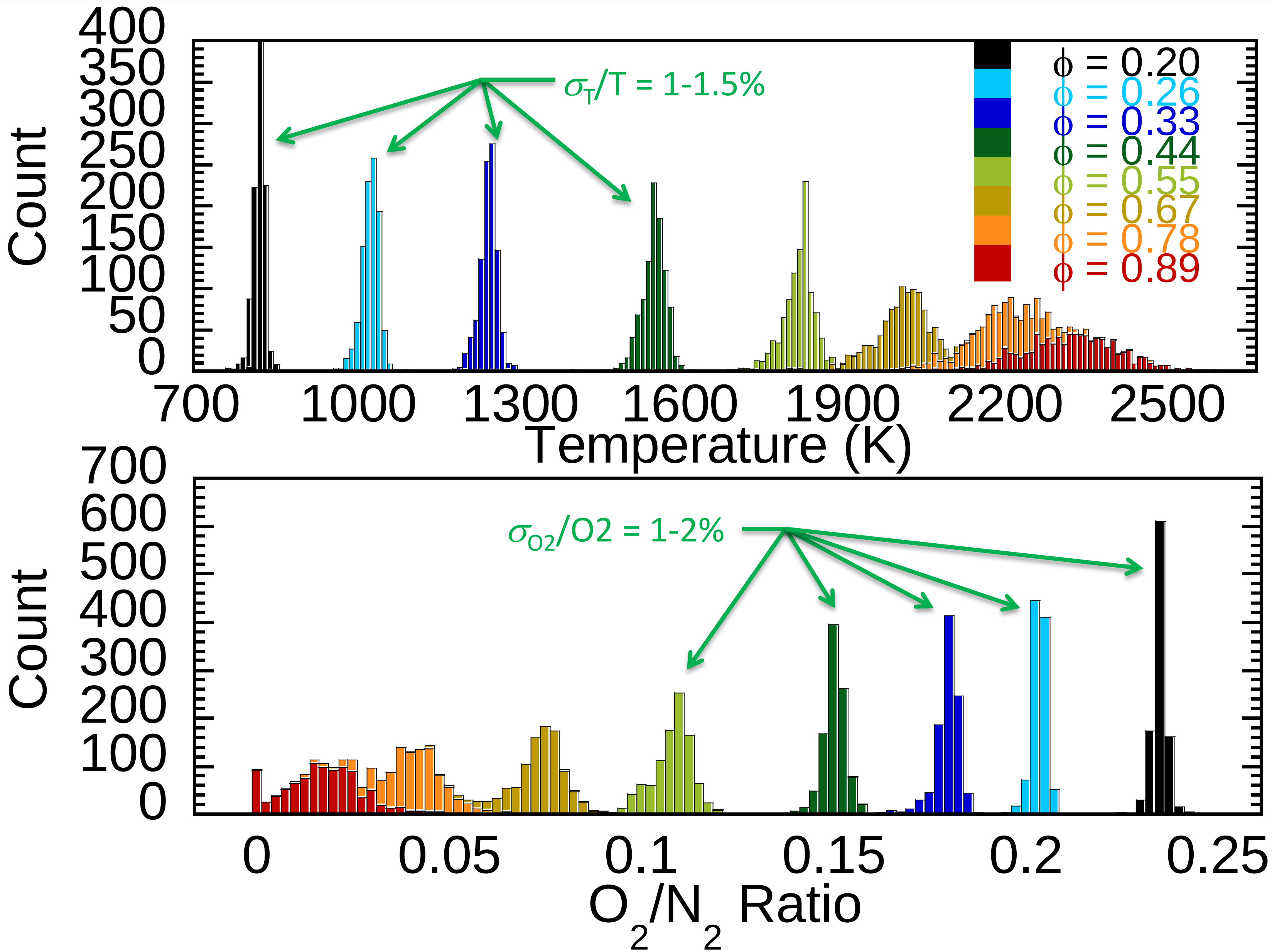
H_2/Air Flame Measurement Accuracy



H_2/Air Flame Measurement Accuracy

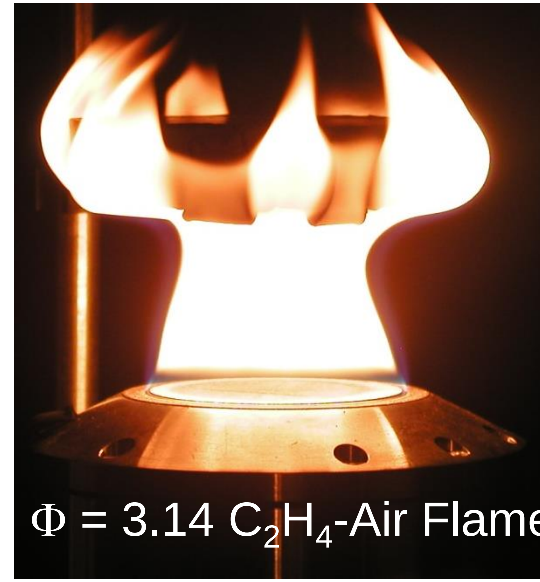


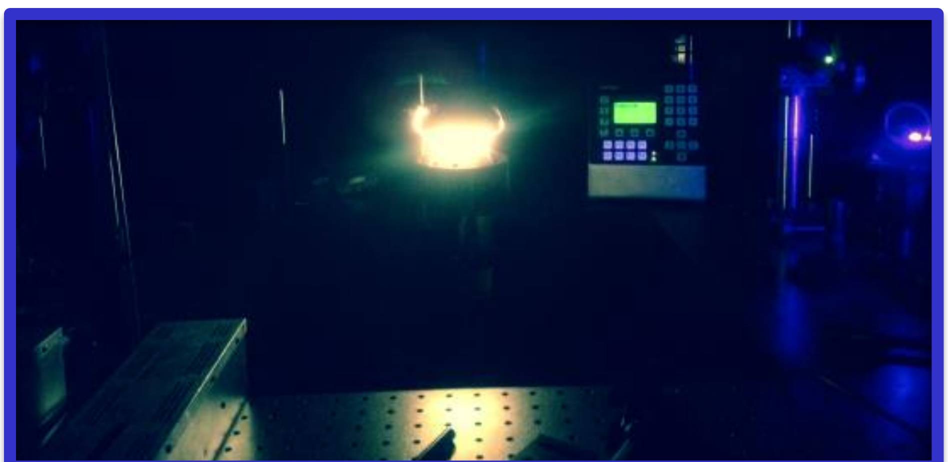
H_2/air Flame Measurement Precision

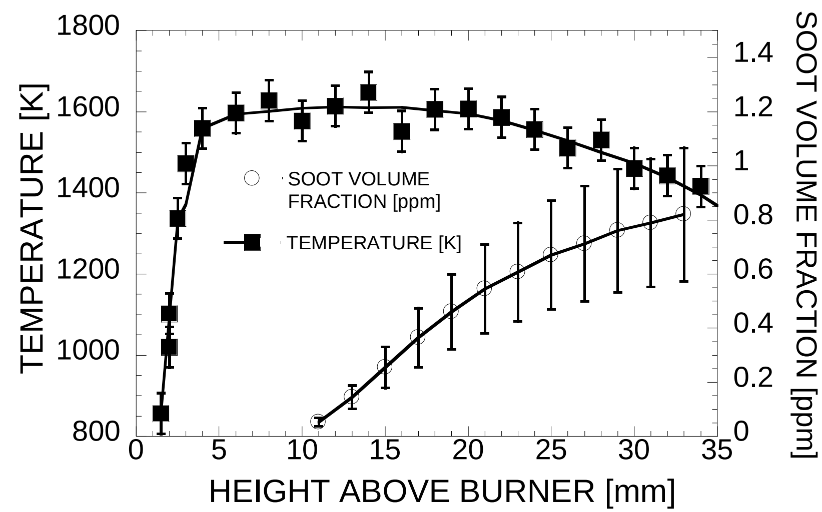


C_2H_4/Air Flames on McKenna Burner

- 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

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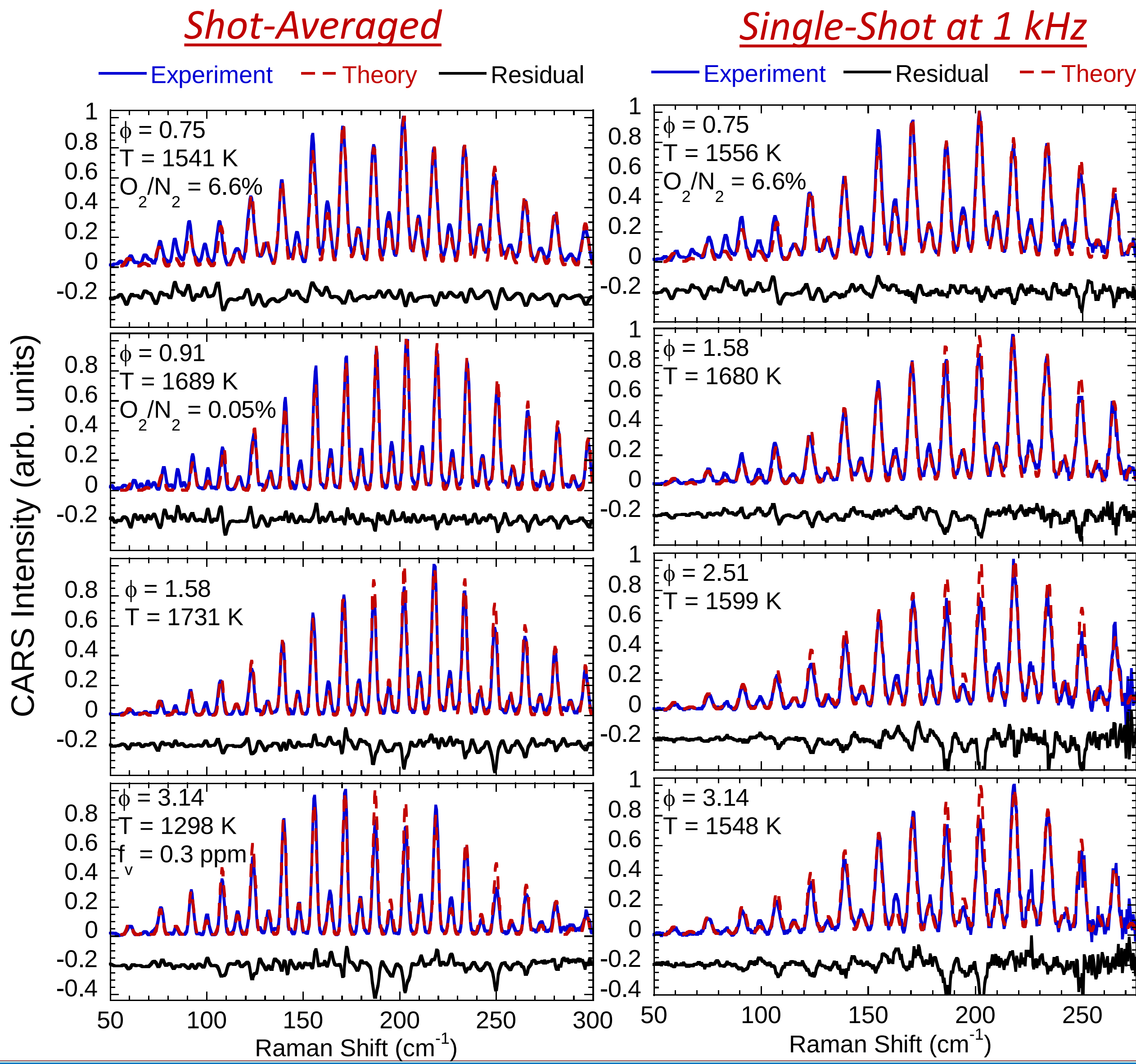
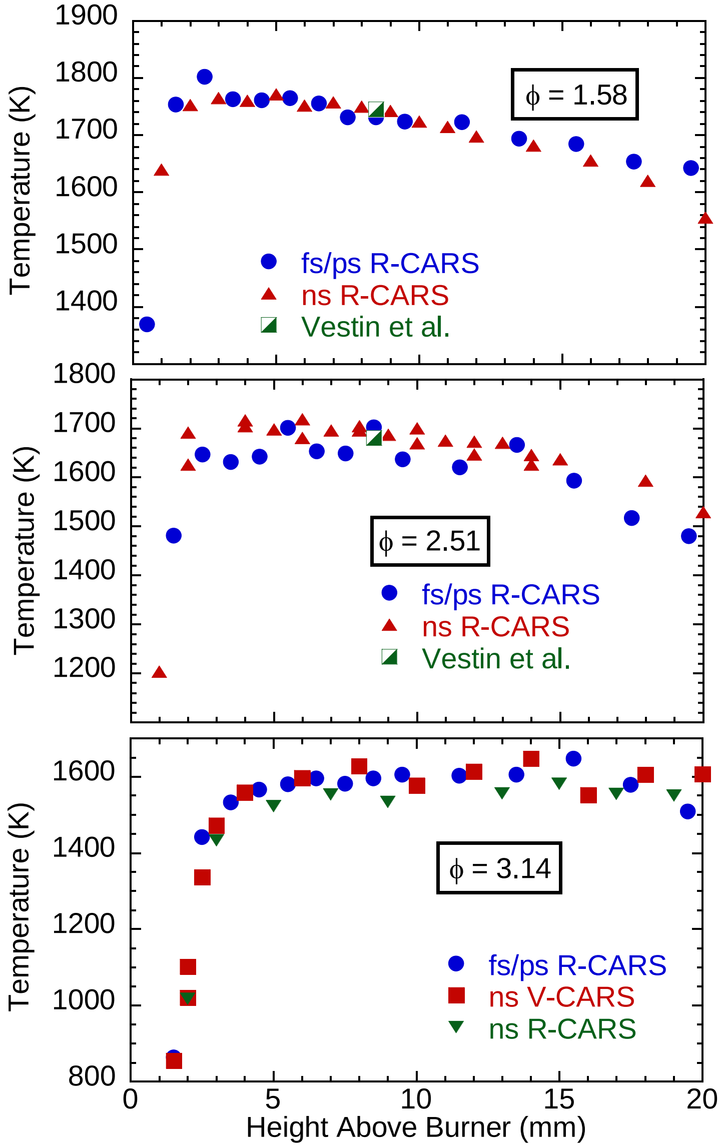


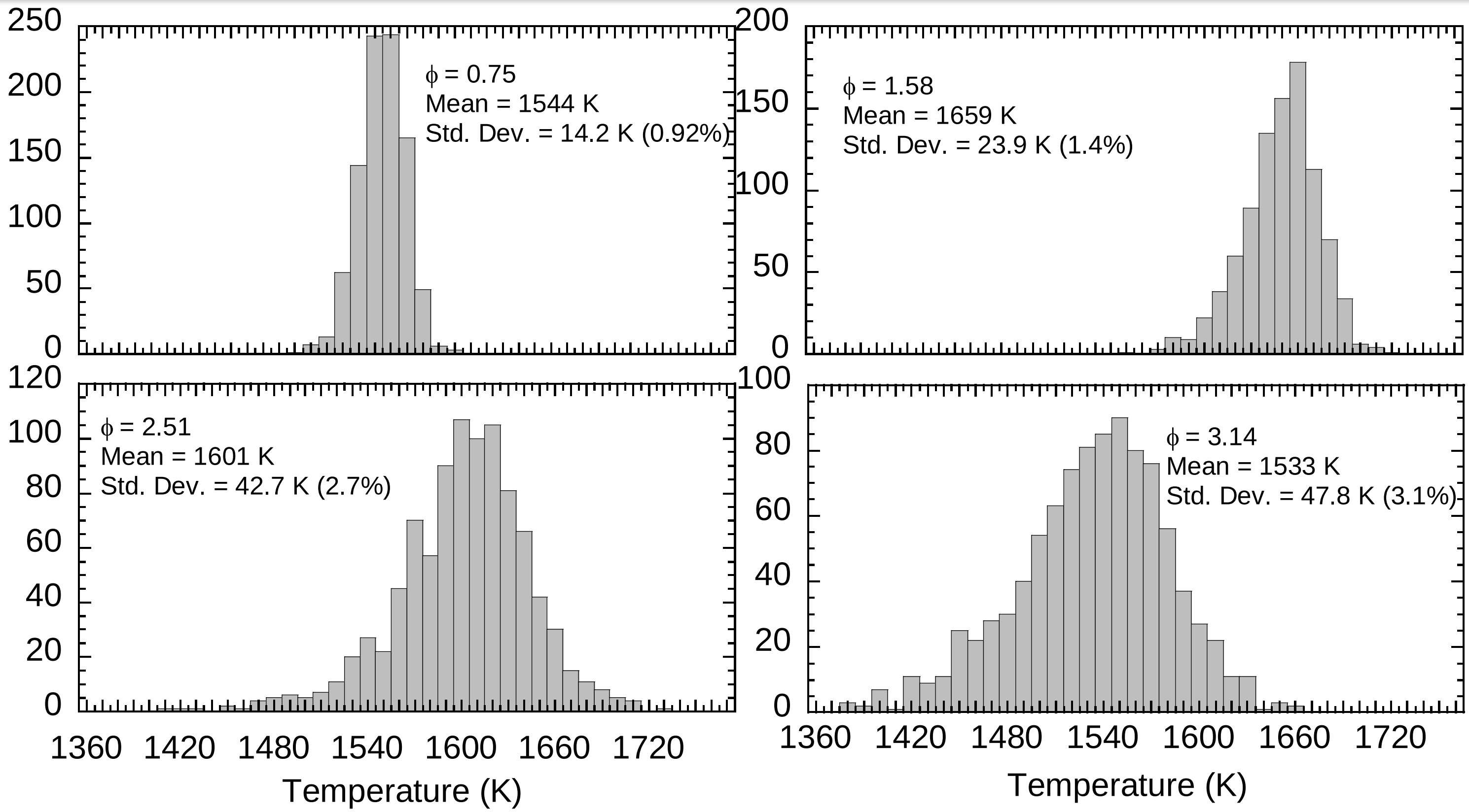
- Spectra acquired for fuel-lean to rich sooting flames
- High-quality fits observed for $\phi < 1$
- Systematic bias toward “underfit” of isolated lines for fuel-rich flames
- Fitted temperature appears to be robust
- Reliable spectra obtained in ***sooting regions*** of the flame
- ***No EM gain used***

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



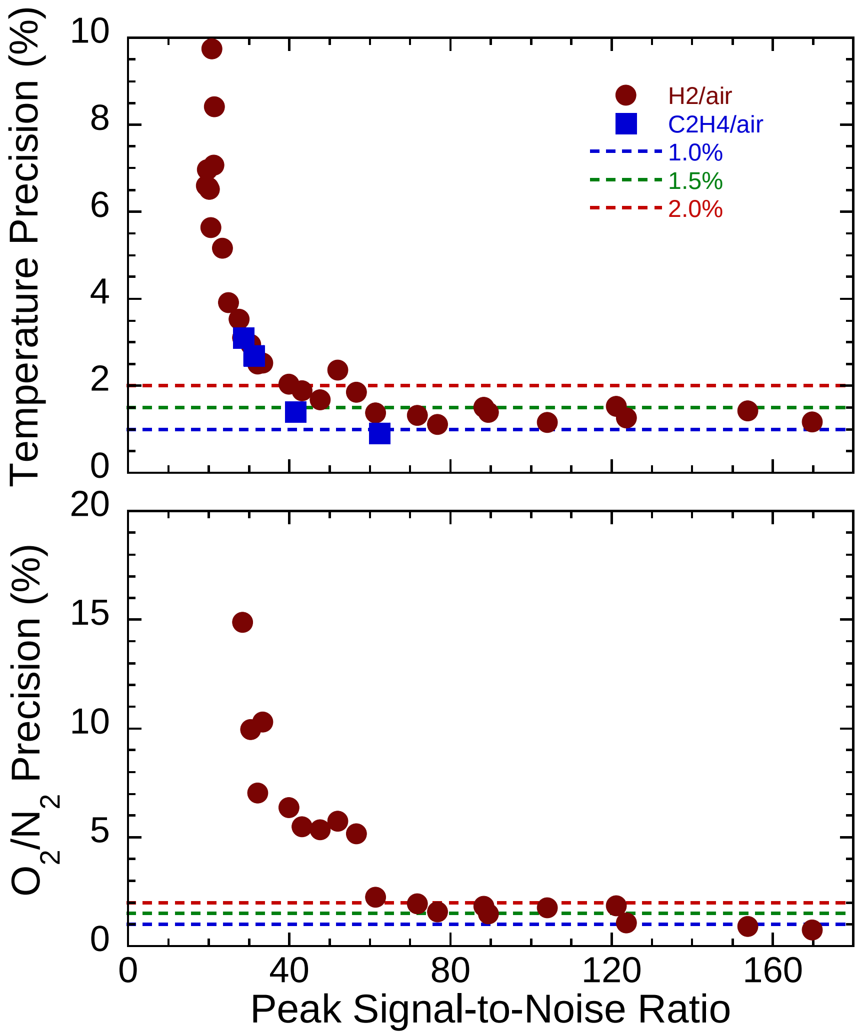
- Comparison is most valid in stable region 5-15 mm height
- ns-CARS temperatures based on fits to shot-averaged spectra
- Agreement of fs/ps and ns CARS is within 1-3% in these ***fuel-rich, sooting*** flames
- Within reported accuracy of rotational ns-CARS
- ns-CARS measurements
  - Rotational CARS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

Temperature Precision in C_2H_4/Air Flames

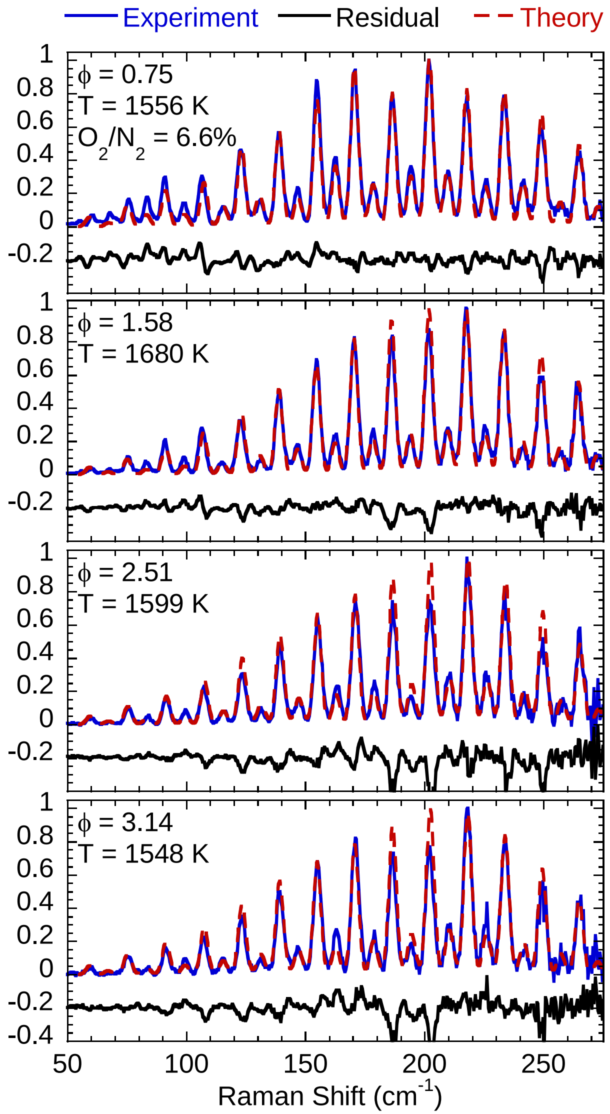


- Measurement volume positioned 11.5 mm above burner ***where flame is stable***
- **Small (8%) variation in temperature in this range of ϕ**
- Precision is 0.9 to 1.4% in leanest flames investigated : ***CARS photon yields are highest***
- Precision degrades to ~3% in richest flames
- Precision appears to be correlated to photon yield

Impact of Signal-to-Noise on Precision



Single-Shot at 1 kHz

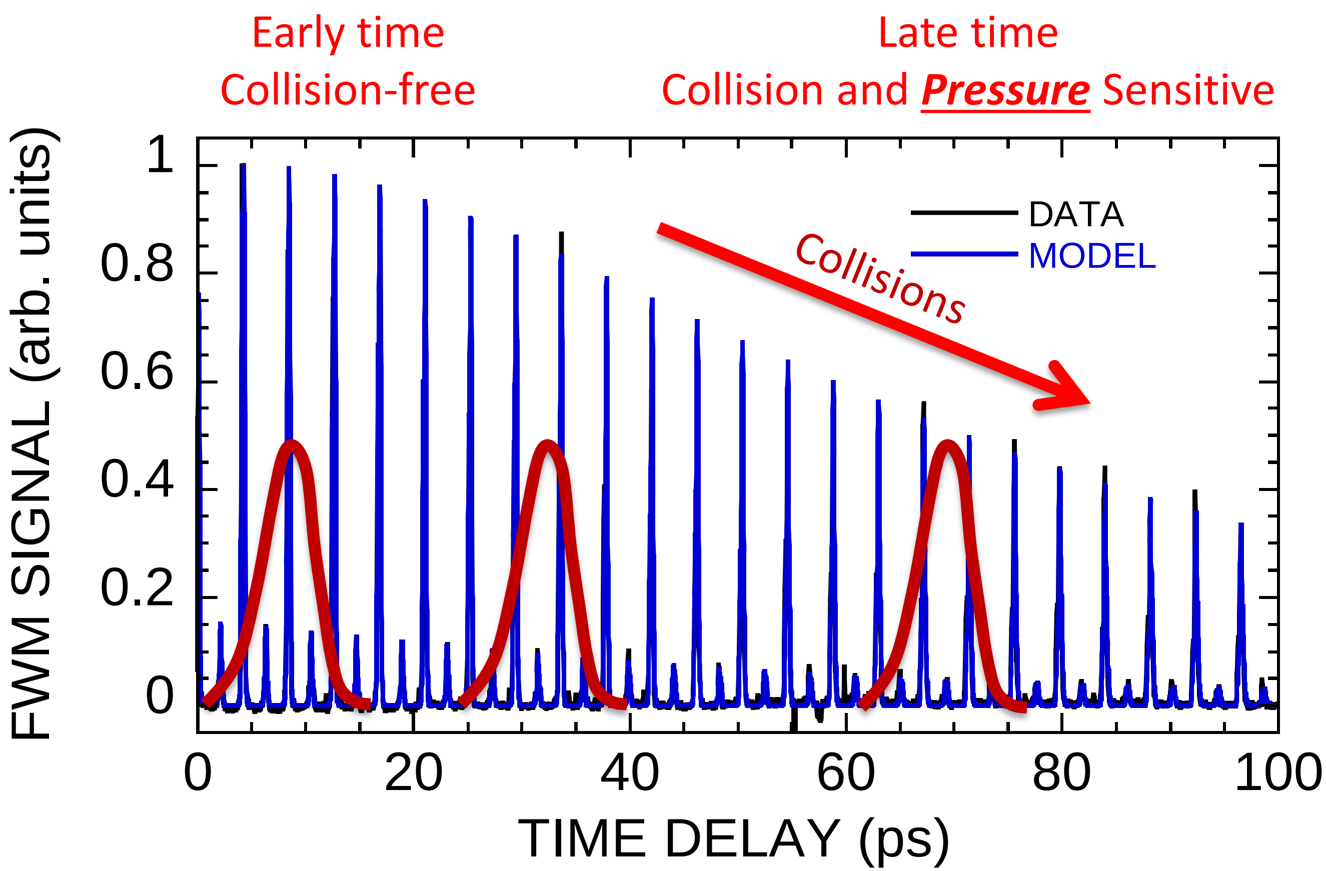


Summary and Conclusions

- 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 $\phi = 0.75$ to 3.14
- Temperature accuracy is typically 3% or better
- Uncertainty in temperature standards is 3%
- Good accuracy in O_2/N_2 ratio observed in H_2/air flames
- Precision is outstanding for temperatures up to 1600-1800 K *if peak SNR is* ~ 40 or more
    - σ_T/T =1-1.5%; σ_{O2}/O2 = 1-2%
    - Periodic oscillations of 30 K (2%) amplitude realized in Hencken burner

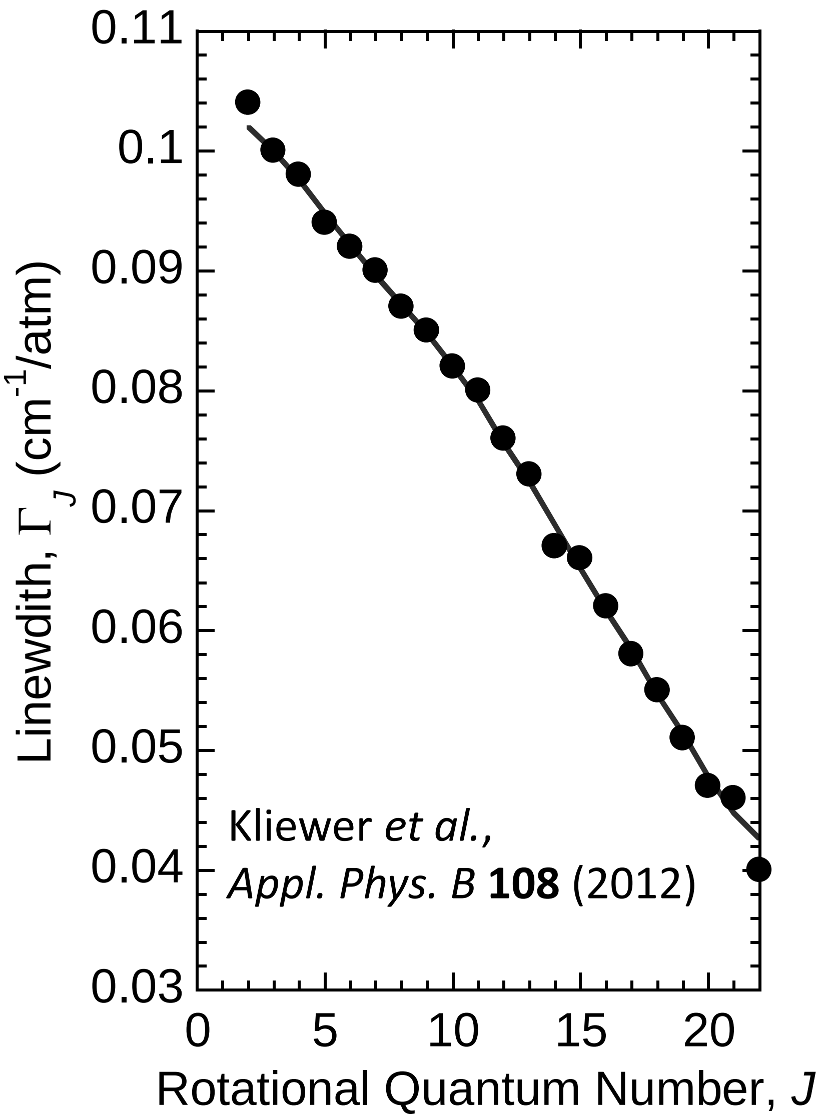
“Bonus Material” Pressure Measurements

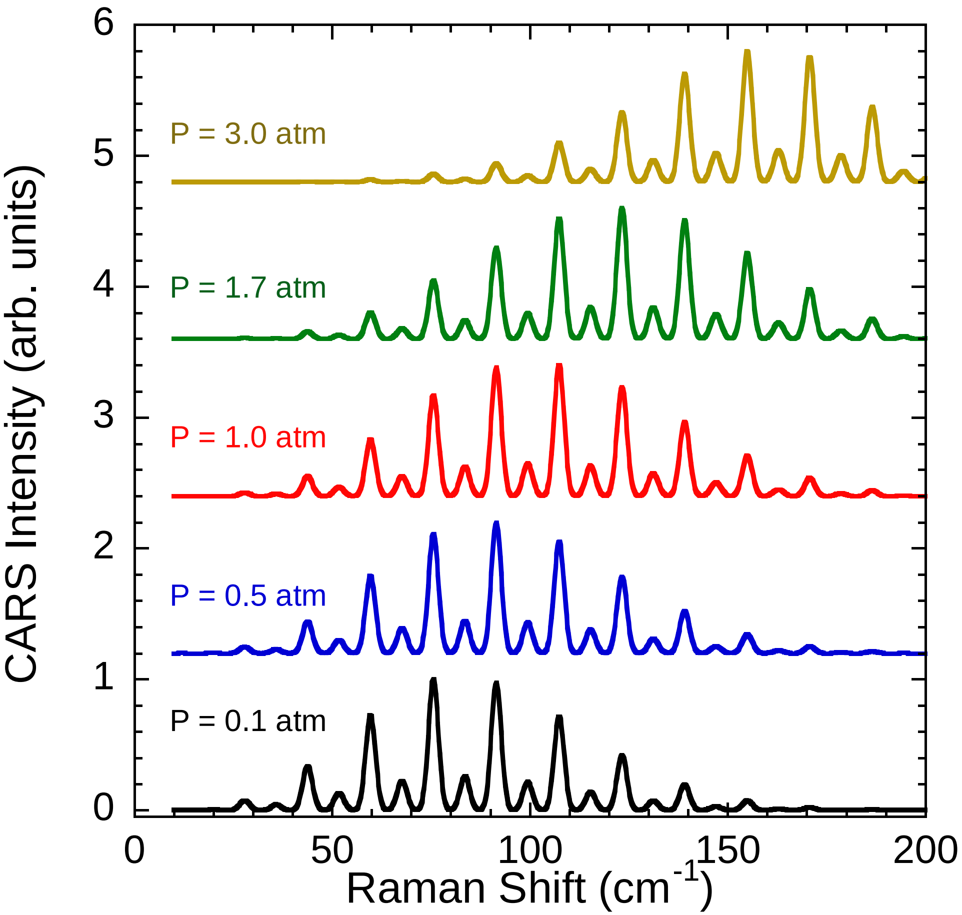
Measured Response in N_2 at T = 300 K



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

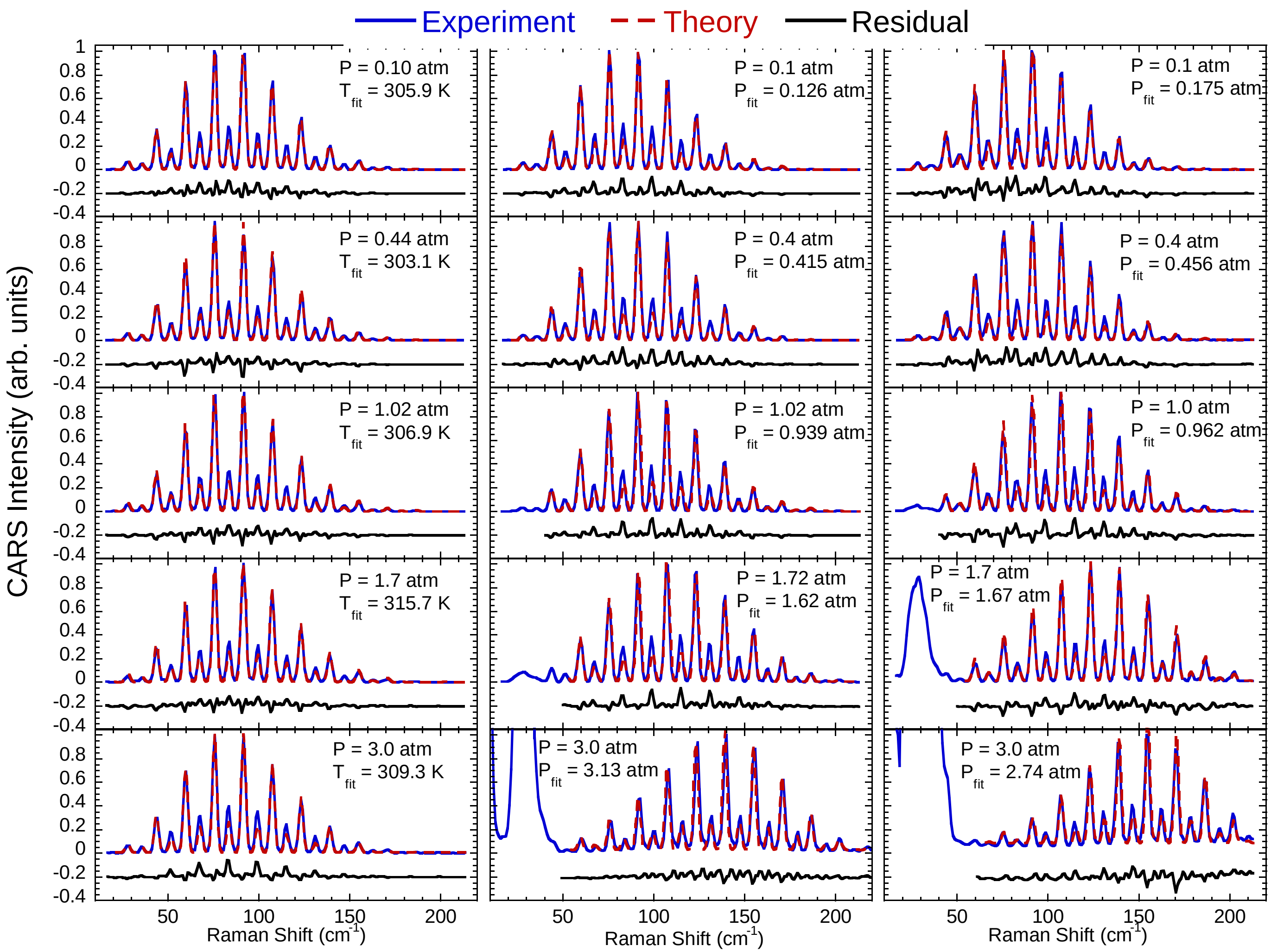
J-dependent dephasing enables pressure measurements





Calculated spectra at T = 294 K
Probe delay τ = 300 ps

N_2 CARS measurements in room-temperature gas cell



***P* = 0.1 atm**

***P* = 0.4 atm**

***P* = 1.0 atm**

***P* = 1.7 atm**

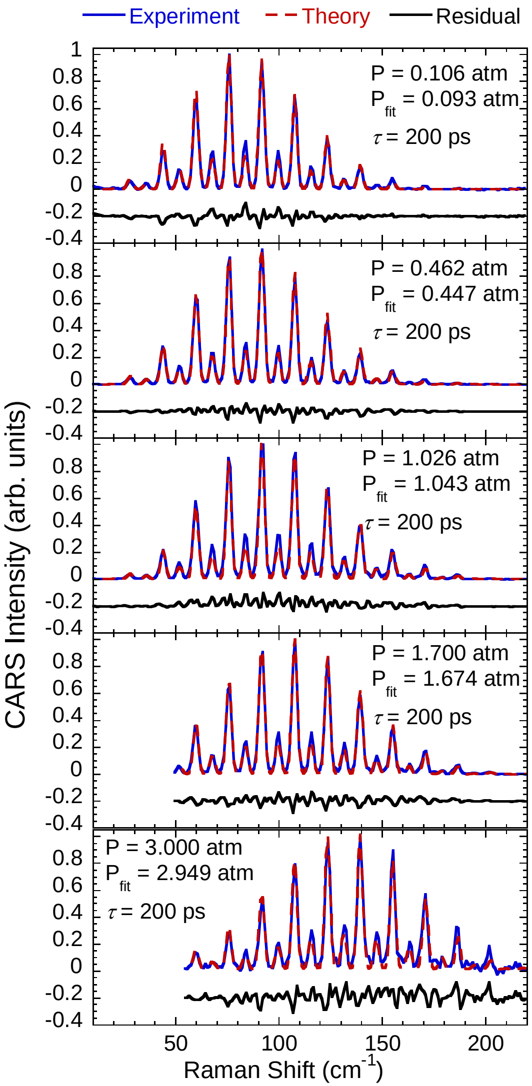
***P* = 3.0 atm**

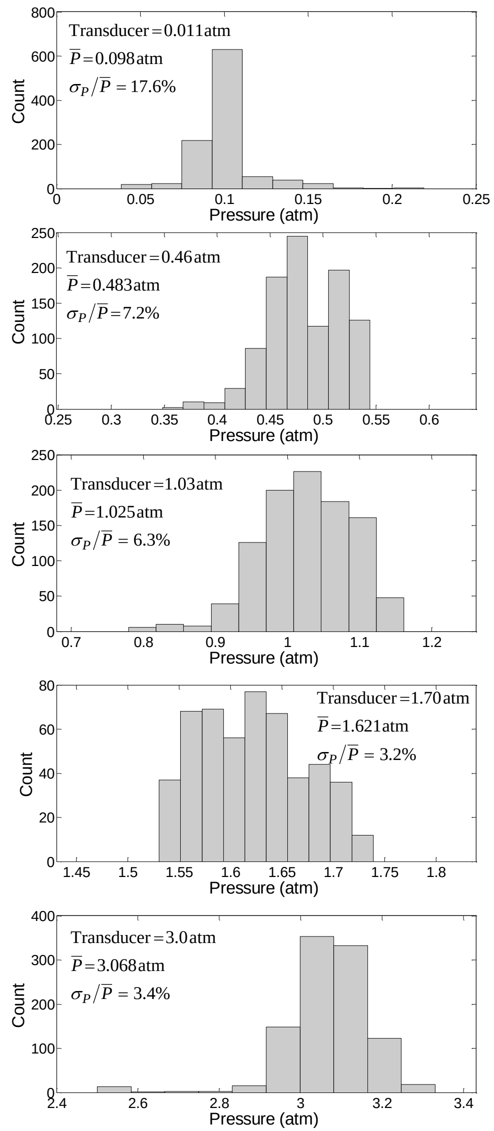
τ = 11.5 ps

τ = 201 ps

τ = 300 ps

Single-Laser-Shot Pressure Measurements

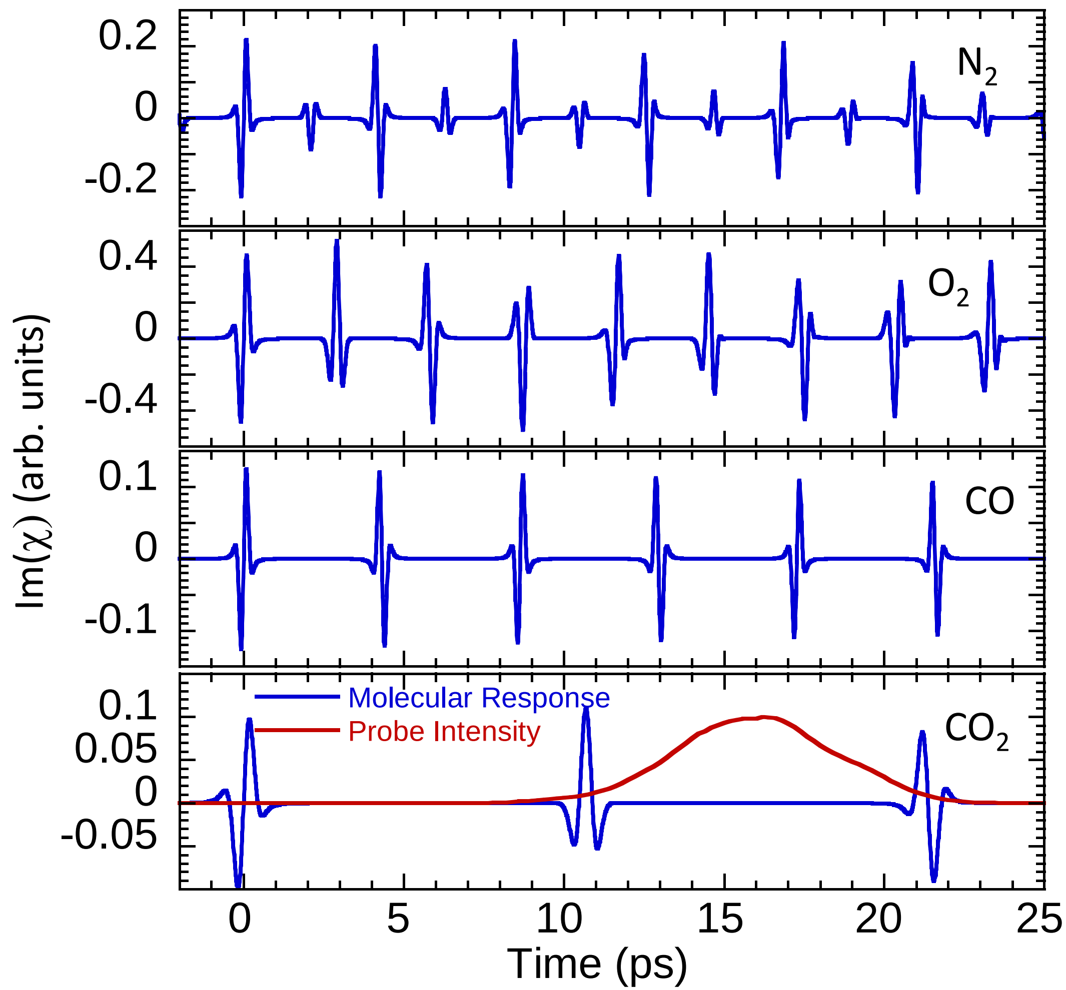




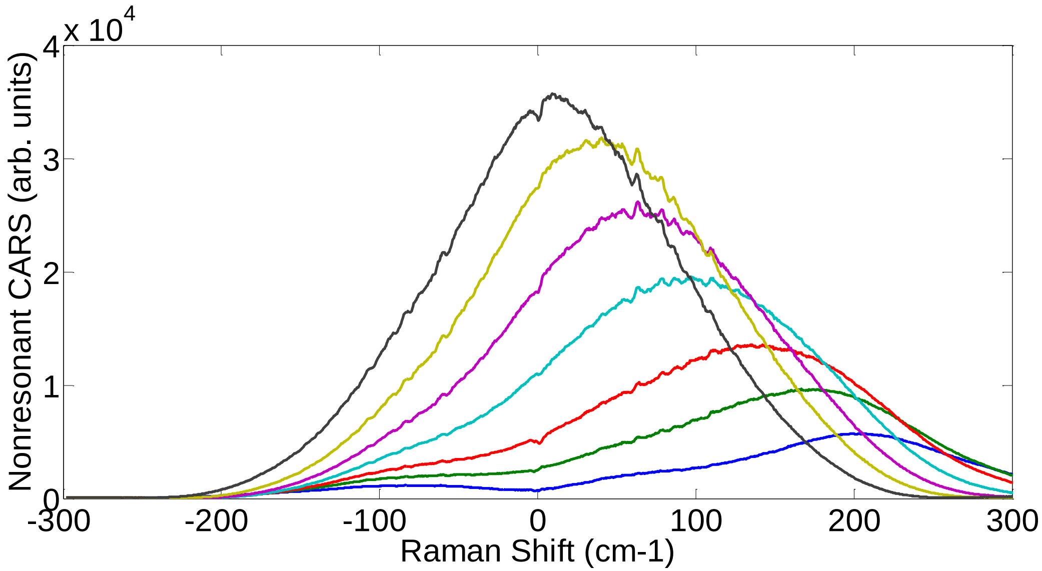
- N_2 in a room-temperature cell
- Long probe delays require high-energy SHBC for good single-shot SNR
- Single-shot means within 5% of transducer or better
- Precision σ_p/P ~ 3-7%
- Sensitivity can be tuned to a given pressure range by adjusting the probe delay
- Improved single-shot precision is possible
- Expect ~2x worse than temperature precision
- Schemes based on two-probe approaches are feasible
- Simultaneous T/P monitoring

Extra Slides for Potential Questions

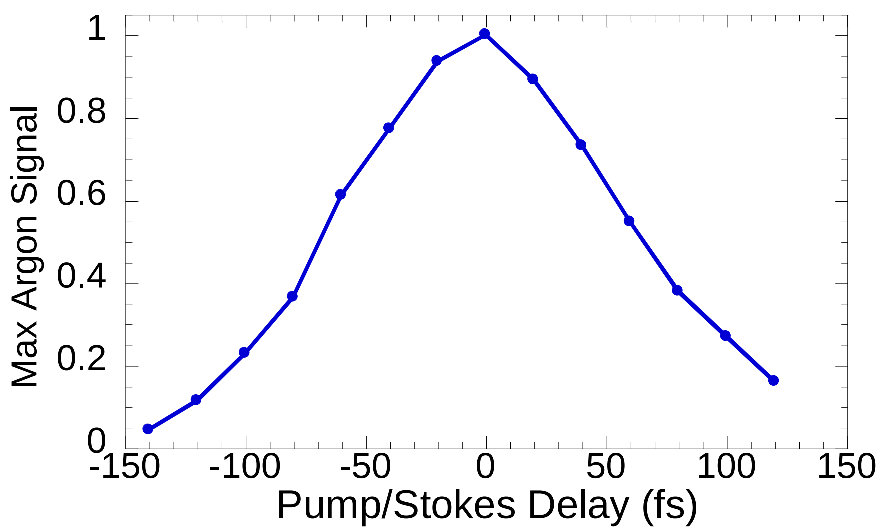
Time Gated Minimization of CO2

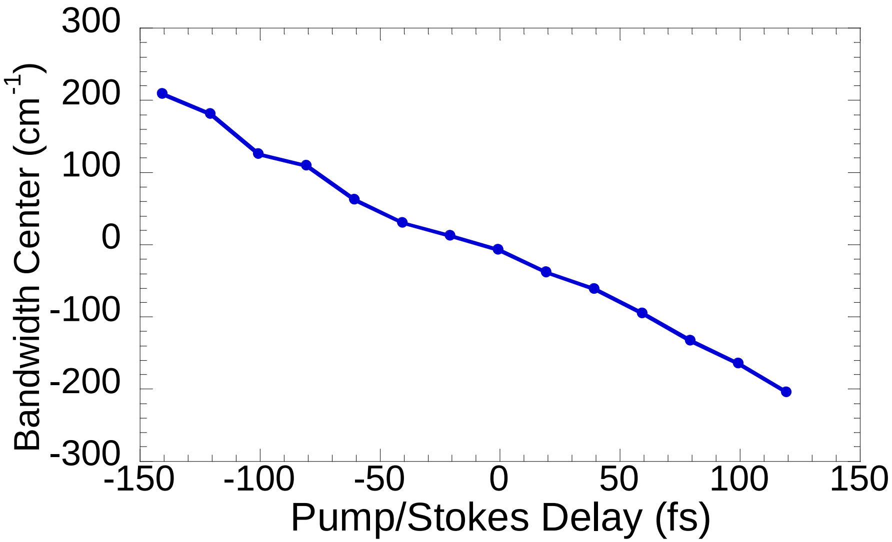


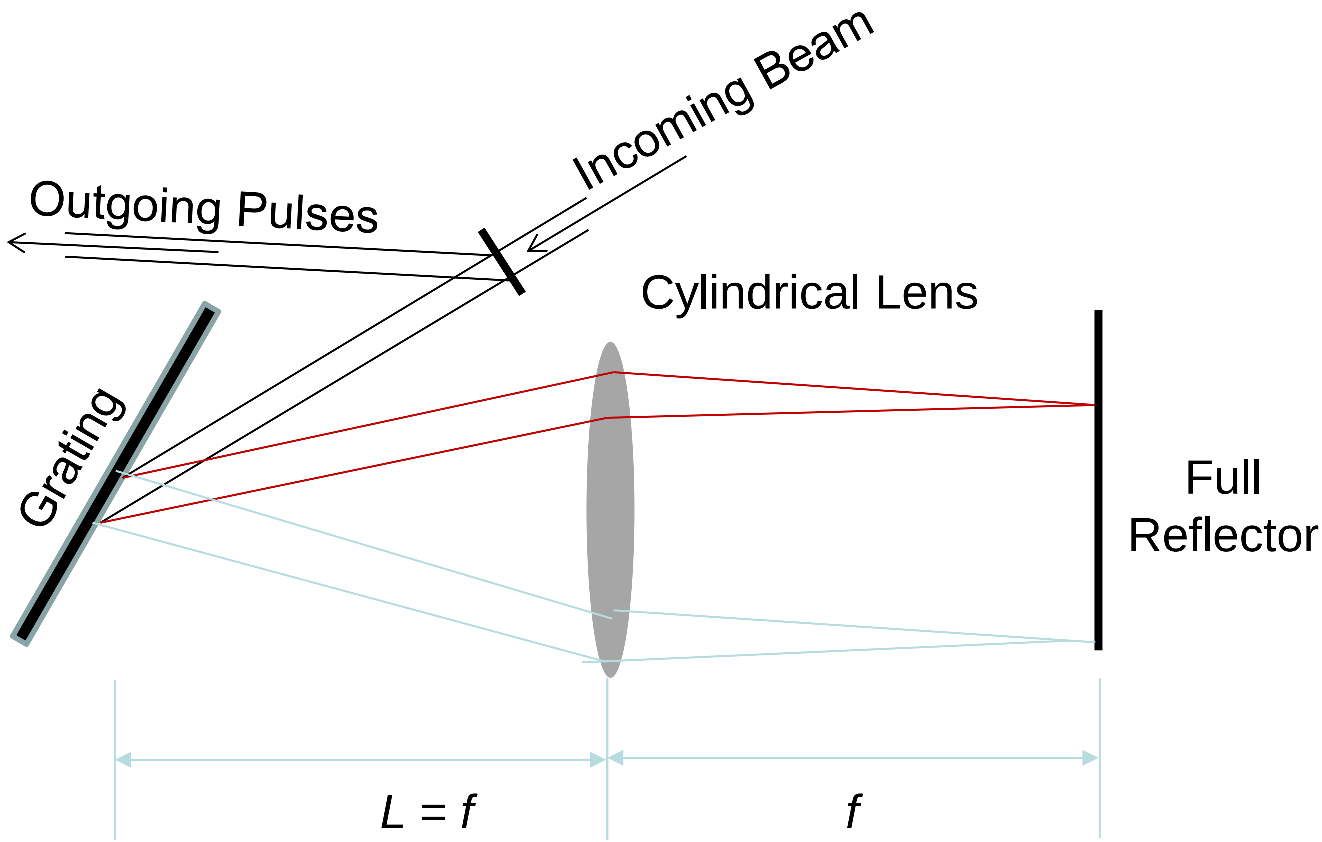
Pump Stokes Delay Shifts Bandwidth Center

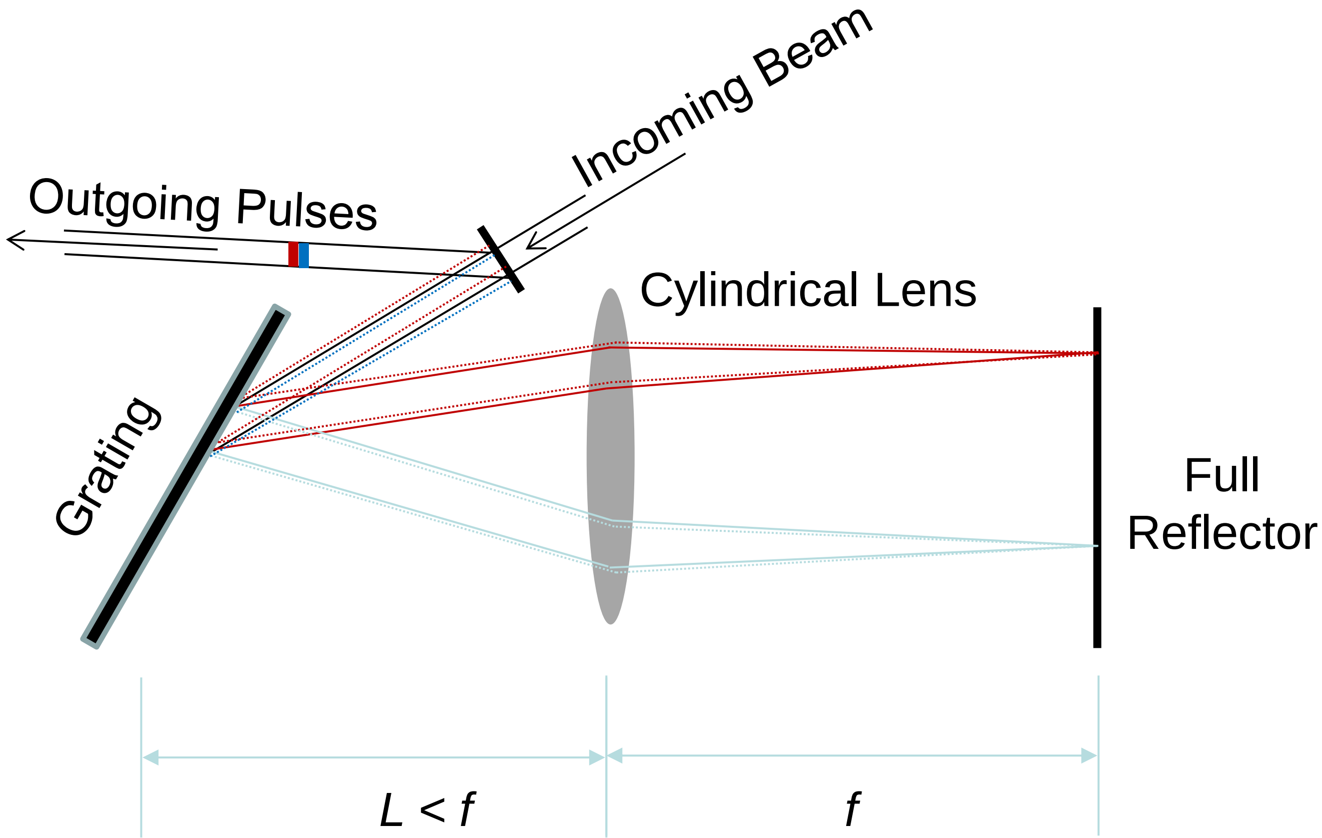


- Delay between pump and Stokes pulses shifts the peak signal
- Allows full utilization of limited bandwidth
- Increased signal for high-J transitions in flames
- 2-3× hit on peak nonresonant signal
- No apparent loss in total bandwidth

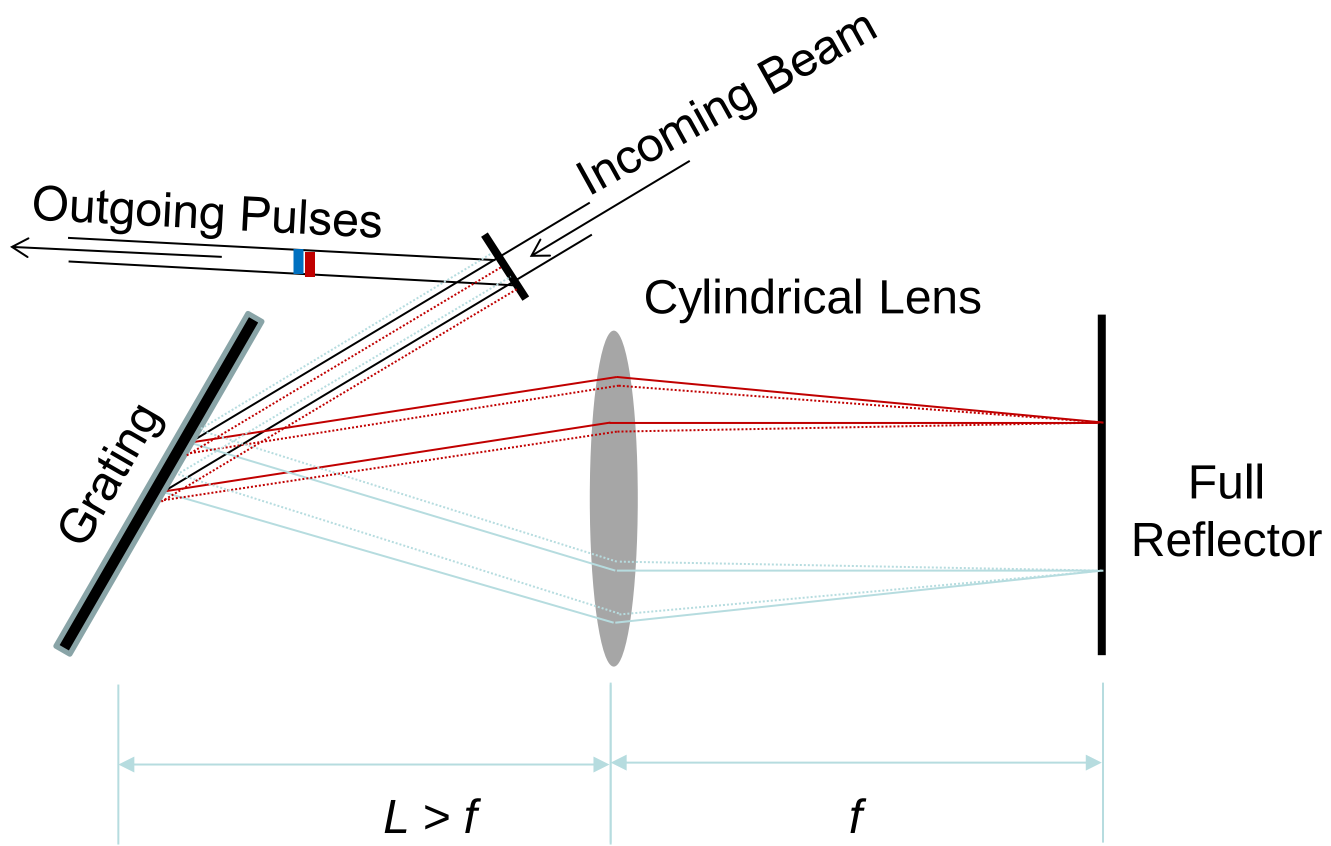




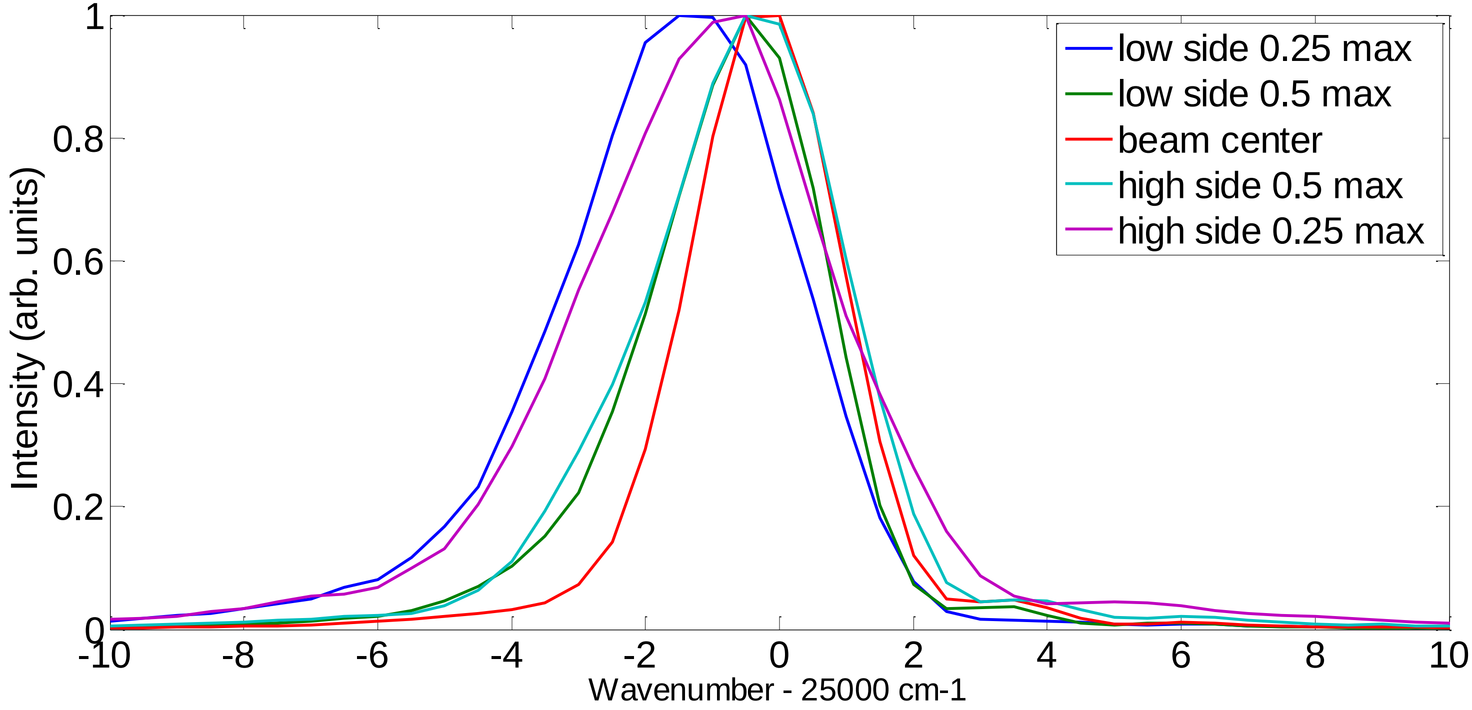

Incoming Beam
Outgoing Pulses
Cylindrical Lens
Grating
Full
Reflector
L = f
f



Incoming Beam
Outgoing Pulses
Cylindrical Lens
Grating
Full
Reflector
L < f
f

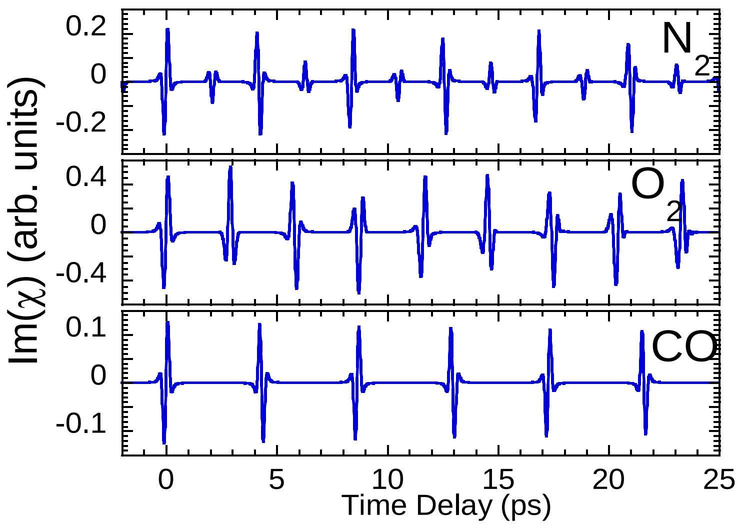

Incoming Beam
Outgoing Pulses
Cylindrical Lens
Grating
Full
Reflector
L > f
f

Vertical Spatial Chirp in SHBC Output



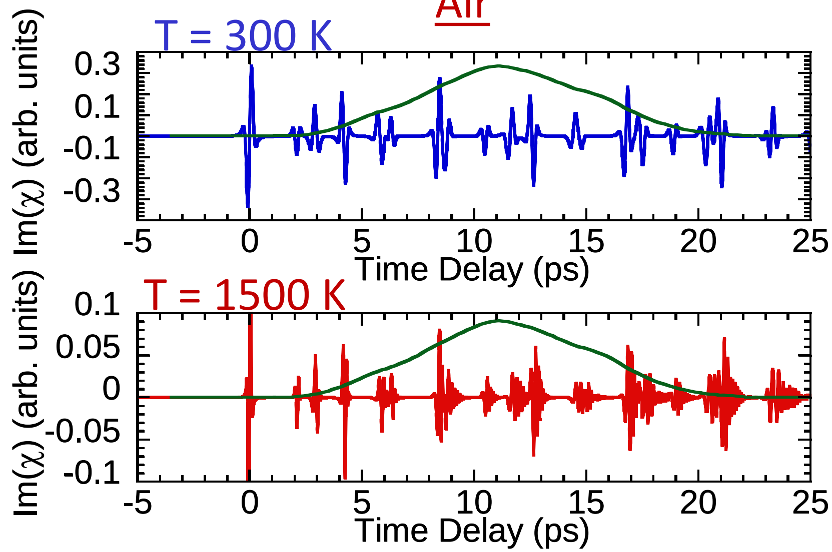
Probe Step and Spectral Synthesis

Pure Gases, T = 300 K

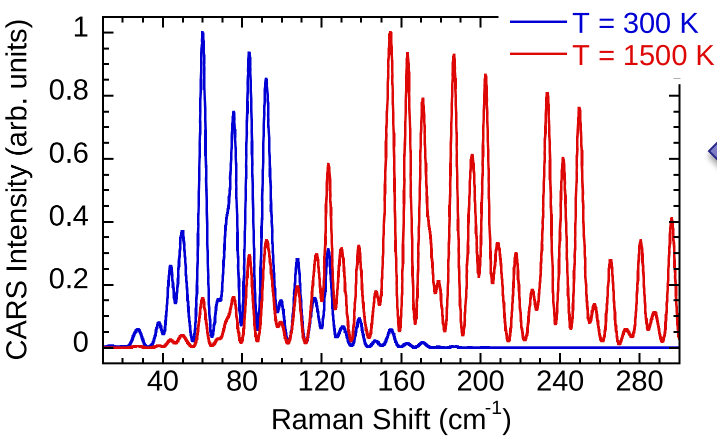


$$\chi = \sum_J W_J \exp\left(i\,\omega_J\, t\right)\exp\left(-\Gamma_J\, t\right)$$

Air



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



FFT

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