Femtosecond Coherent Anti-Stokes Raman Scattering: Recent Applications and the Surprisingly Beneficial Effects of Moderate Pump and Stokes Chirp

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Acknowledgments

- Levi Thomas (PhD 2017, now an Assistant Professor at Air Force Institute of Technology), Mingming Gu (PhD Student), Dr. Aman Satija (Research Scientist), Ziqiao Chang (Undergraduate Researcher).

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Outline of the Presentation

- Introduction and Motivation
- Raman Scattering and Coherent Anti-Stokes Raman Scattering (CARS) Spectroscopy
- Chirped-Probe-Pulse (CPP) Fs CARS Measurements of Temperature in Flames: Frequency Spread Dephasing
- CPP FS CARS Measurements in Spray and Sooting Flames
- Effects of Moderate Pump/Stokes Chirp
- Conclusions and Future Work

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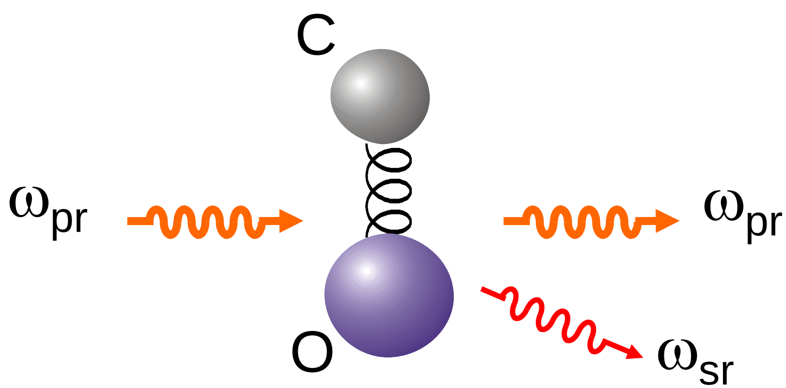
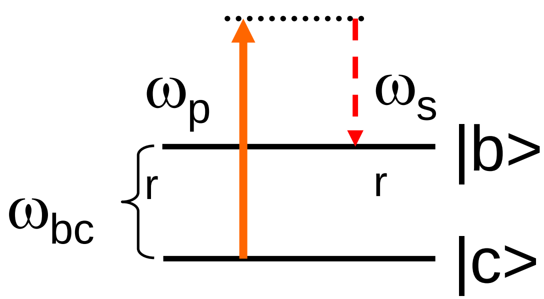
Raman Scattering

Raman effect:

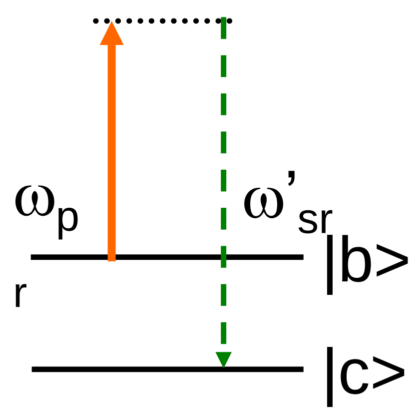


Chandrasekhara Venkata Raman (1888-1970)

1930 Nobel Prize in Physics



Stokes scattering



anti-Stokes scattering

Leonid Isaakovich Mandelshtam (1879-1944)

He called it “combination light scattering“.

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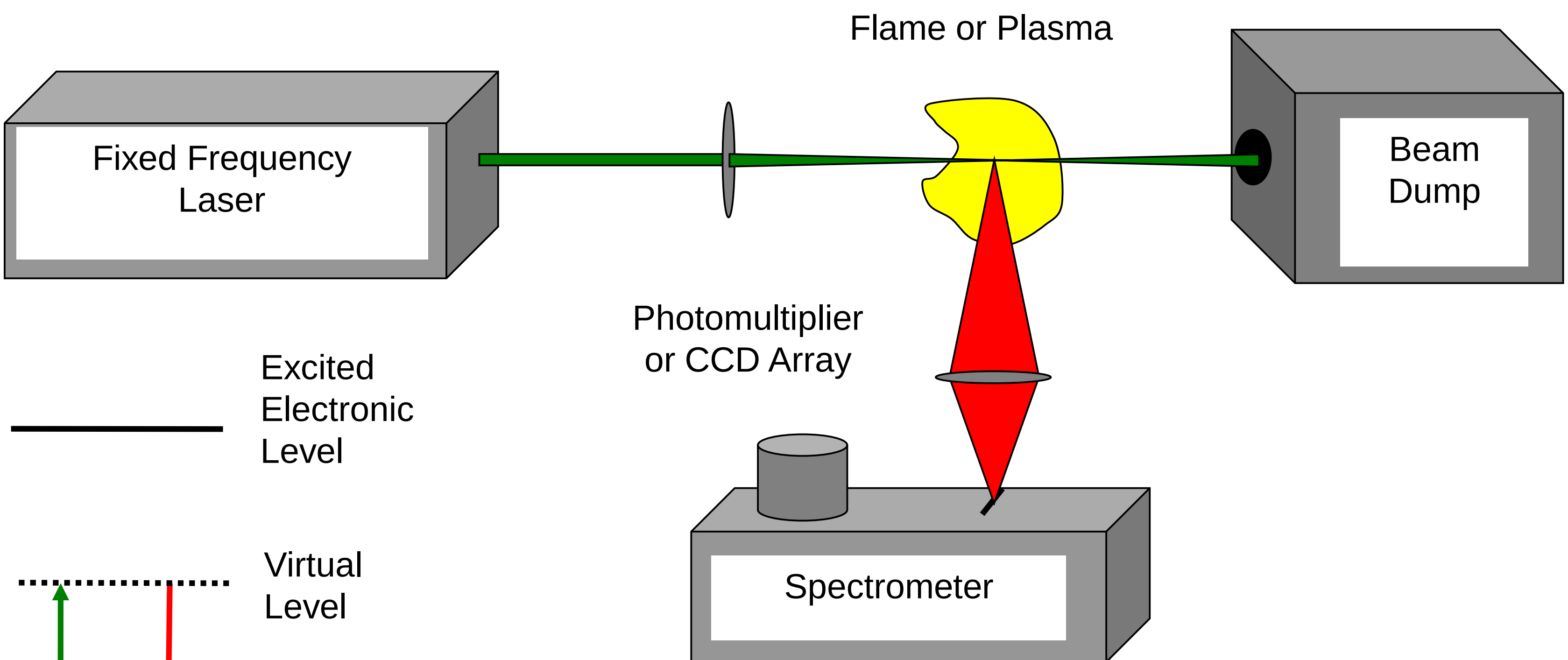
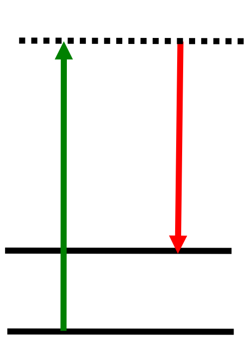
Slides courtesy of Alexei Sokolov

Raman Scattering





Species-specific, spatially resolved, quantitative, weak signal, applicable only for clean flames

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Raman Measurements in High-Pressure, Lean Direct-Injection CH_4 Flames

Kojima and Nguyen, AIAA Journal, Vol. 46, 3116 (2008)

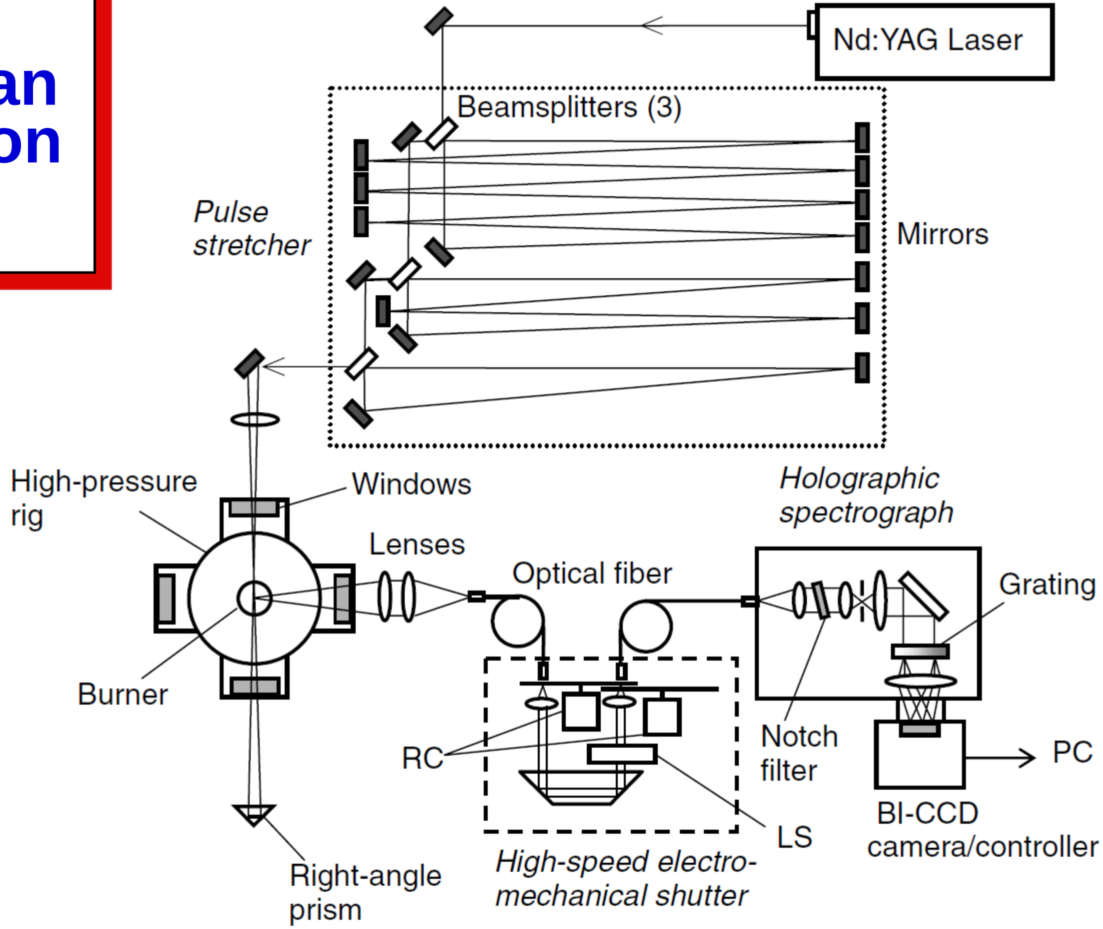

Fig. 2 Schematic of high-performance Raman scattering apparatus. There are four main components to the system: frequency-doubled Nd: YAG pulsed laser, pulse stretcher, high-speed electromechanical shutter, and spectrograph/backside-illuminated CCD camera. Legend—RC: rotary chopper motor/blade combination; LS: leaf shutter; PC: personal computer.

Raman Measurements in High-Pressure, Lean Direct-Injection CH_4 Flames

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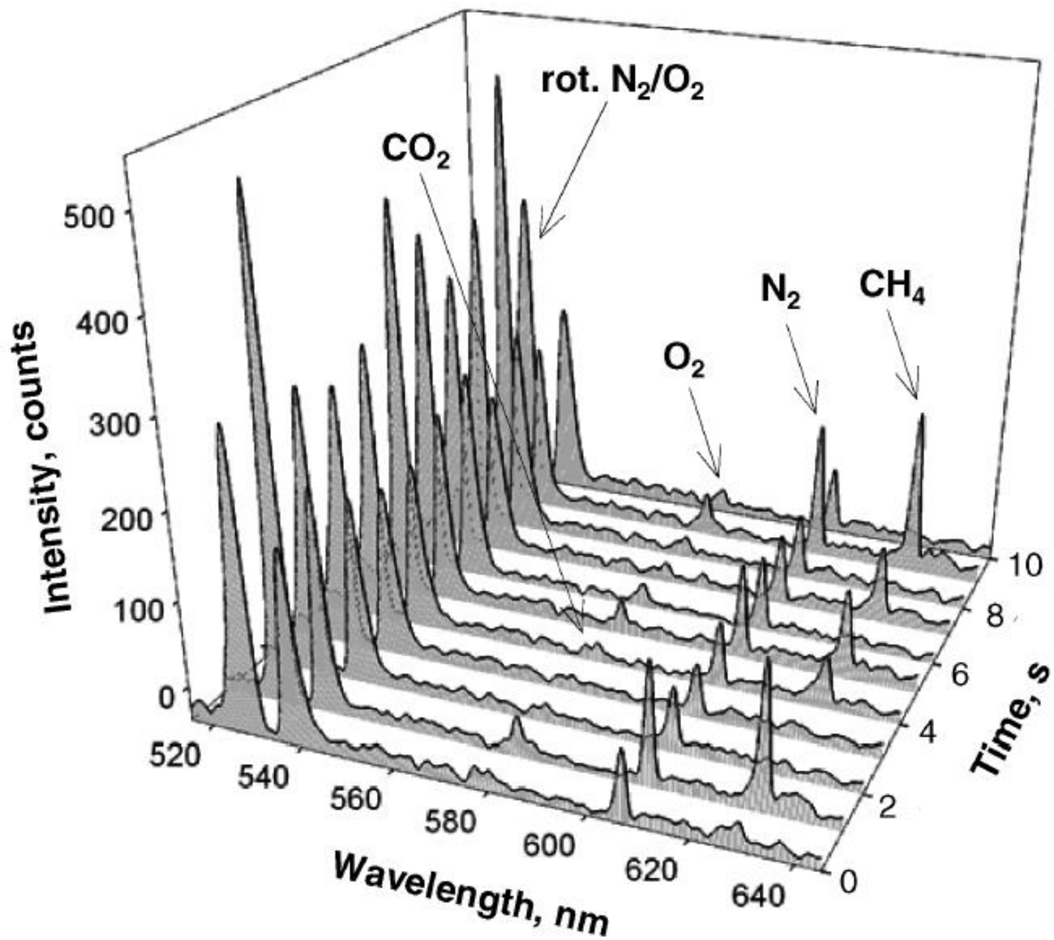
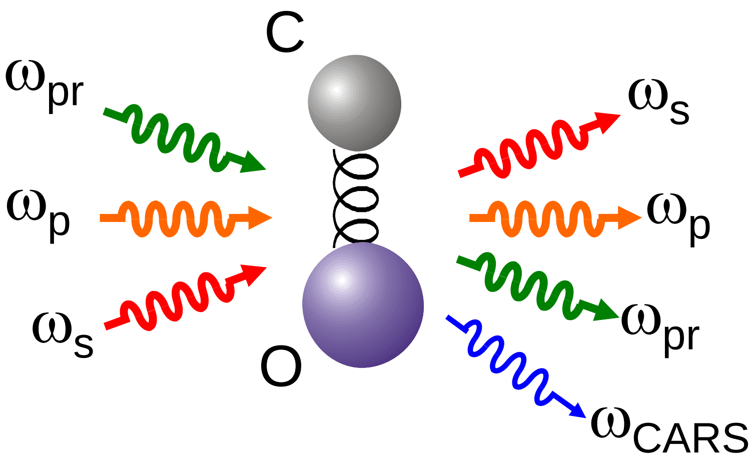


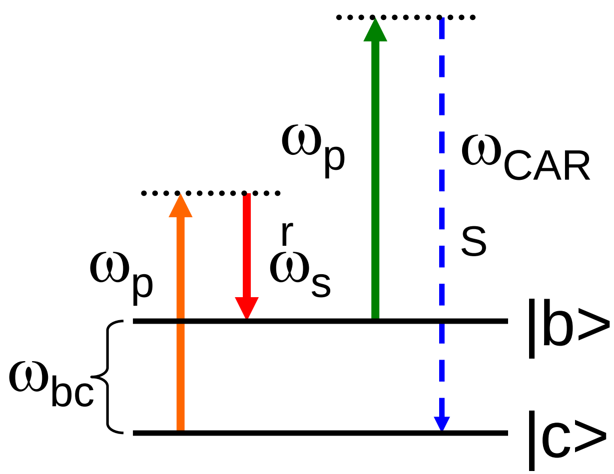
Fig. 3 Typical single-shot Raman spectra (532 nm excitation) measured in the turbulent methane–air flame. Eleven shots are shown out of a total 400 shots obtained by the measurement at $(r, x) = (10, 9)$.

Coherent Anti-Stokes Raman Scattering

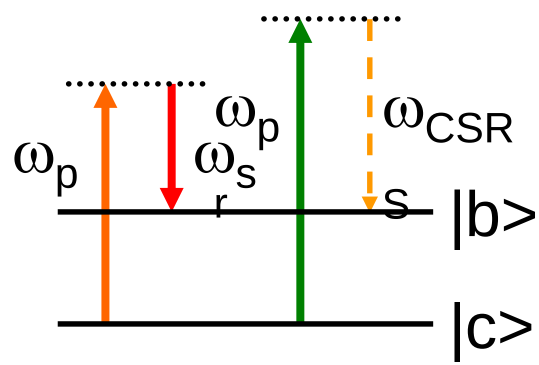


Interesting facts:

- The CARS phenomenon was first reported in 1965 by P. D. Maker and R. W. Terhune, two researchers of the Scientific Laboratory at the Ford Motor Company.
- The name coherent anti-Stokes Raman spectroscopy was assigned almost ten years later, by Begley et al. at Stanford University in 1974.
- CARS first used for gas-phase diagnostics by J.-P. Taran of ONERA in 1973.



CARS



CSRS

Slides courtesy of Alexei Sokolov

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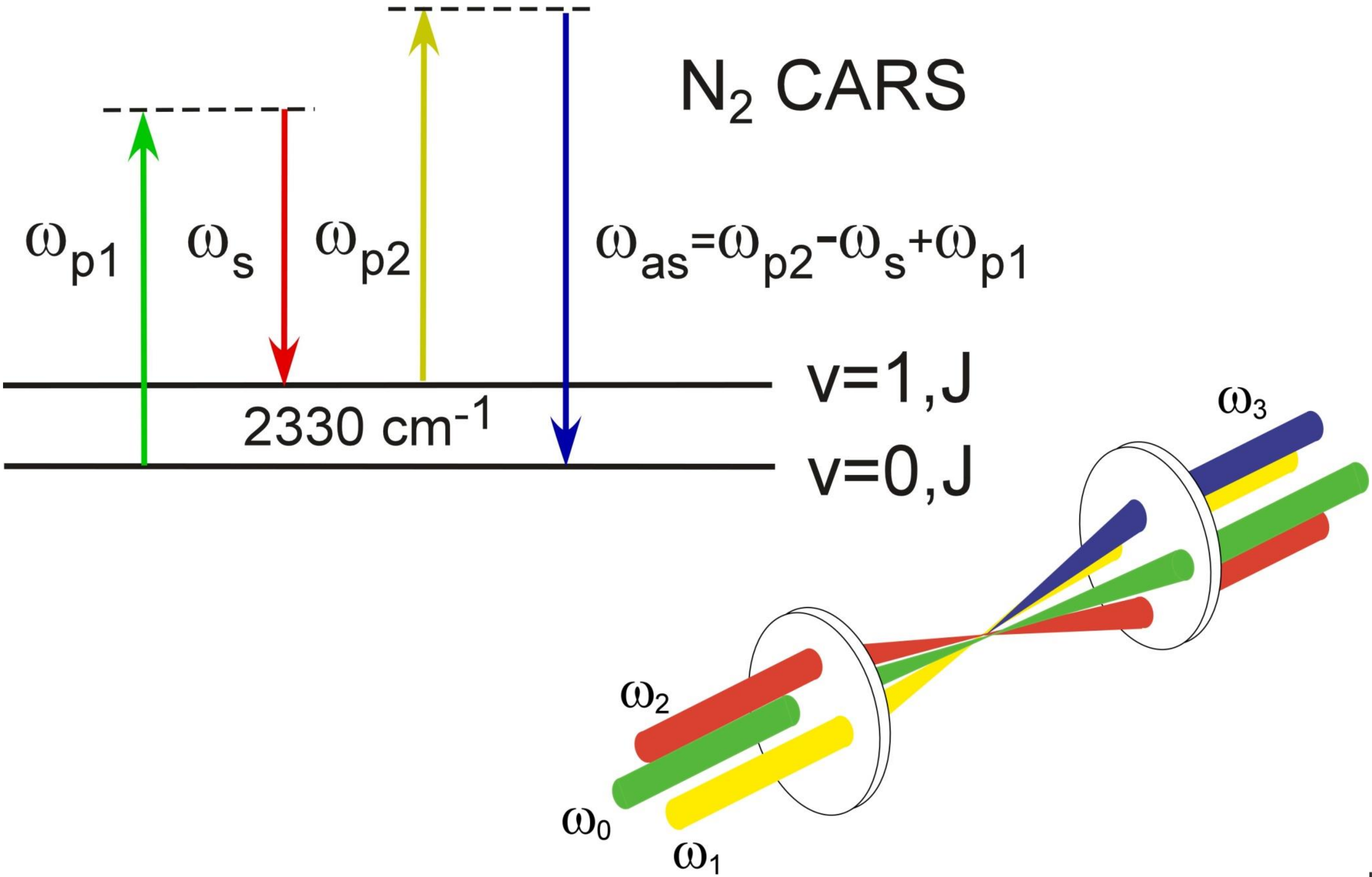
Ns CARS for Gas-Phase Diagnostics



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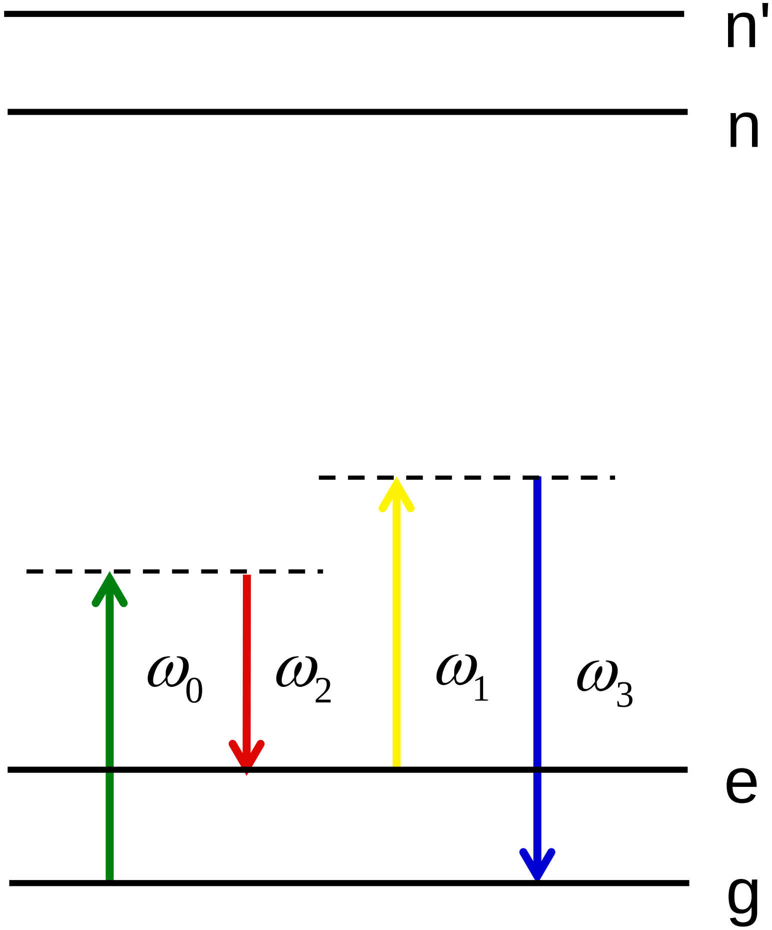
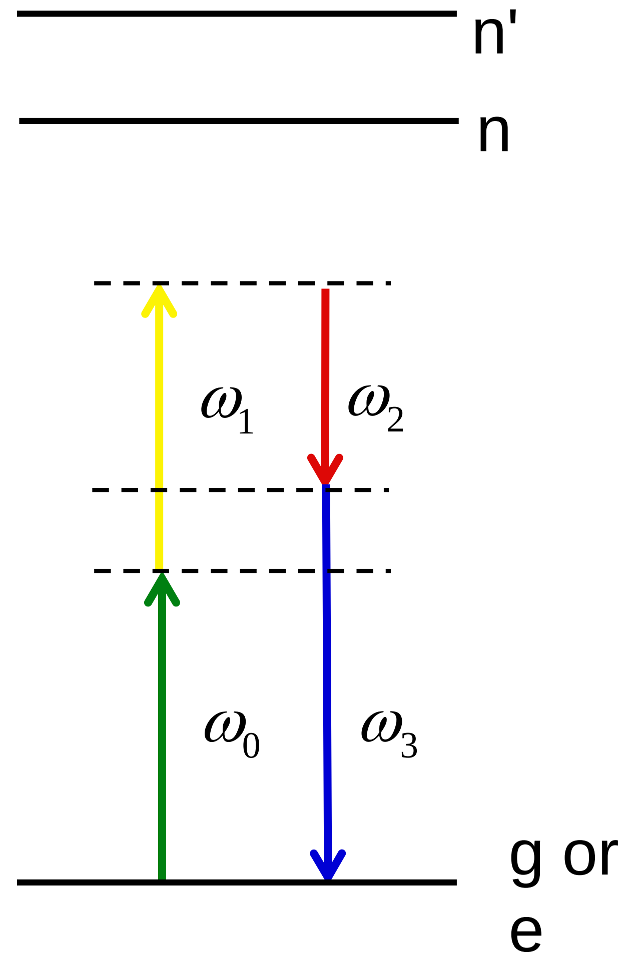
Nonresonant Background for CARS



CARS

$$\omega_{CARS} = \omega_3 = \omega_0 + \omega_1 - \omega_2$$



Nonresonant Four-Wave Mixing

$$\omega_{NR} = \omega_3 = \omega_{CARS}$$

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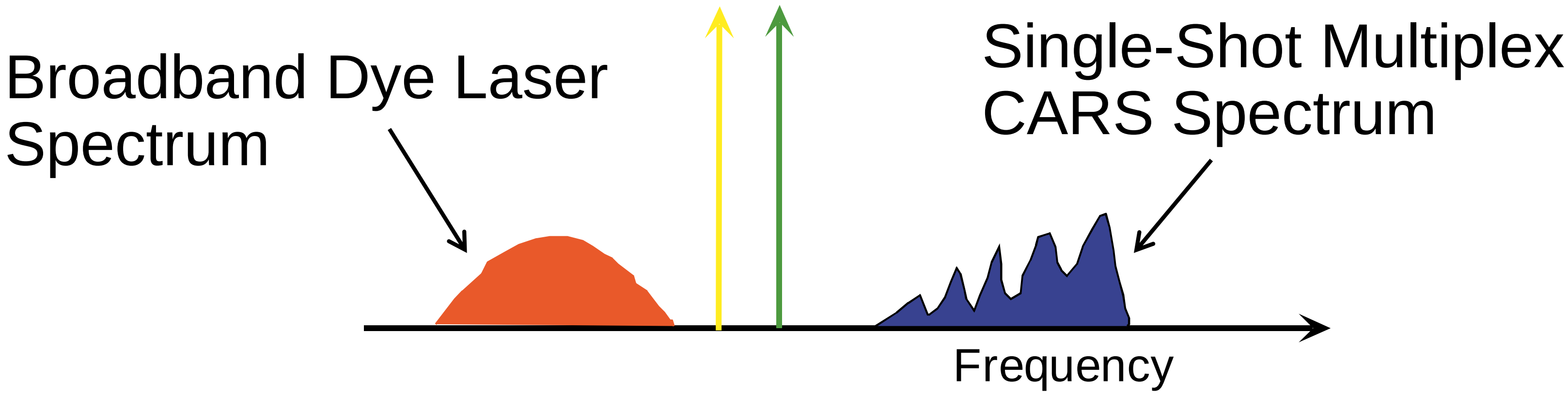
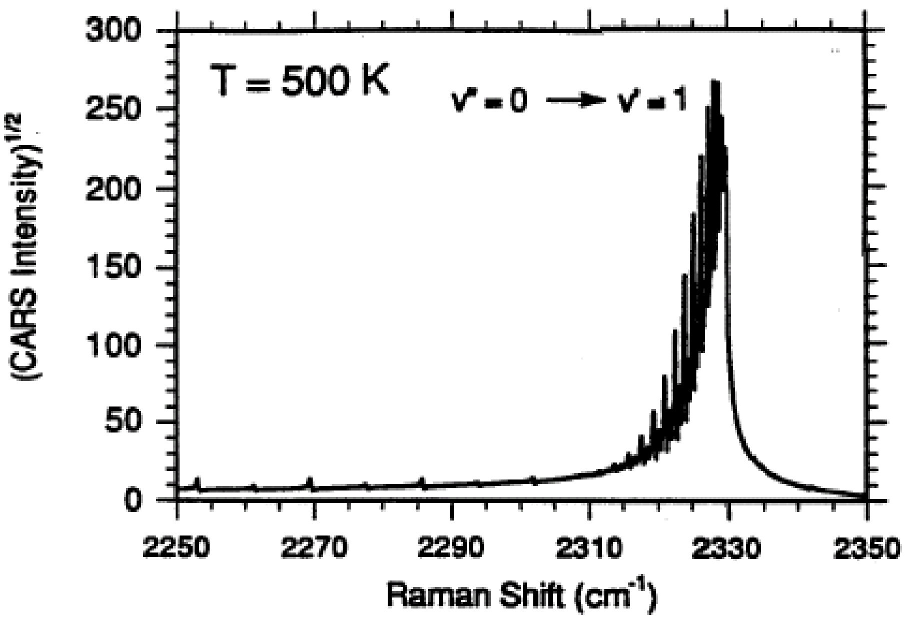
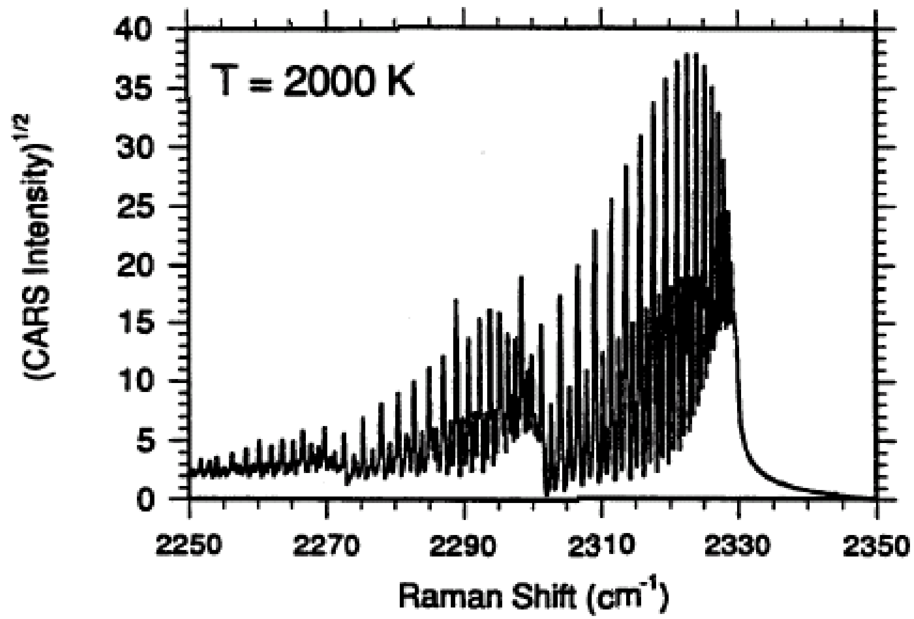
Ns CARS for Gas-Phase Diagnostics







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Ns CARS for Gas-Phase Diagnostics

- Nanosecond CARS using (typically) a Q-switched Nd:YAG laser and broadband dye laser is a well-established technique for combustion and plasma diagnostics
- Actively developed in 1980's for combustion after Alan Eckbreth at UTRC tried to make Raman scattering measurements in combustor primary zone, discovered massive interference due to laser heating of soot particles to 10,000K – now called laser-induced incandescence

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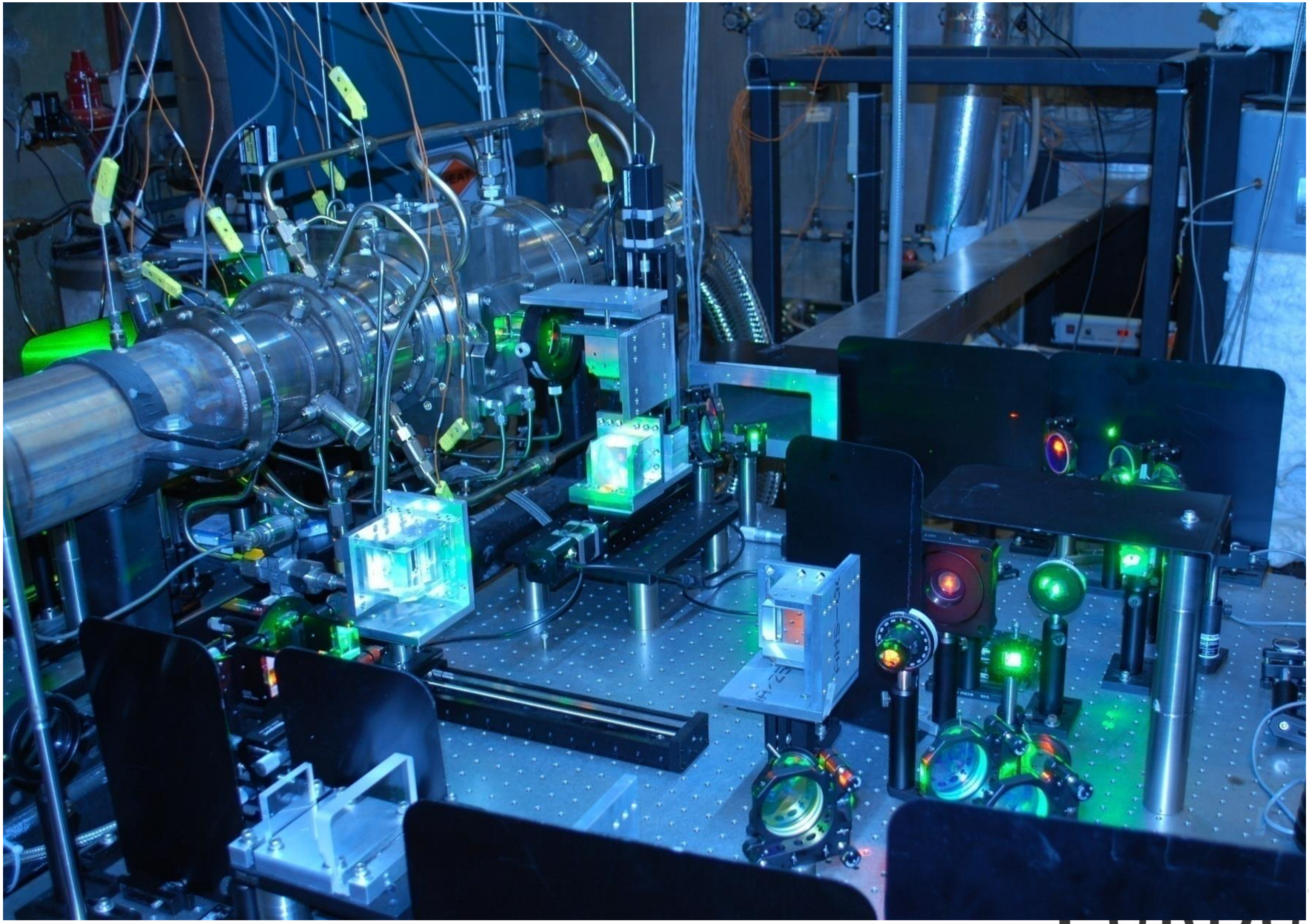
Ns CARS in Gas Turbine Combustor



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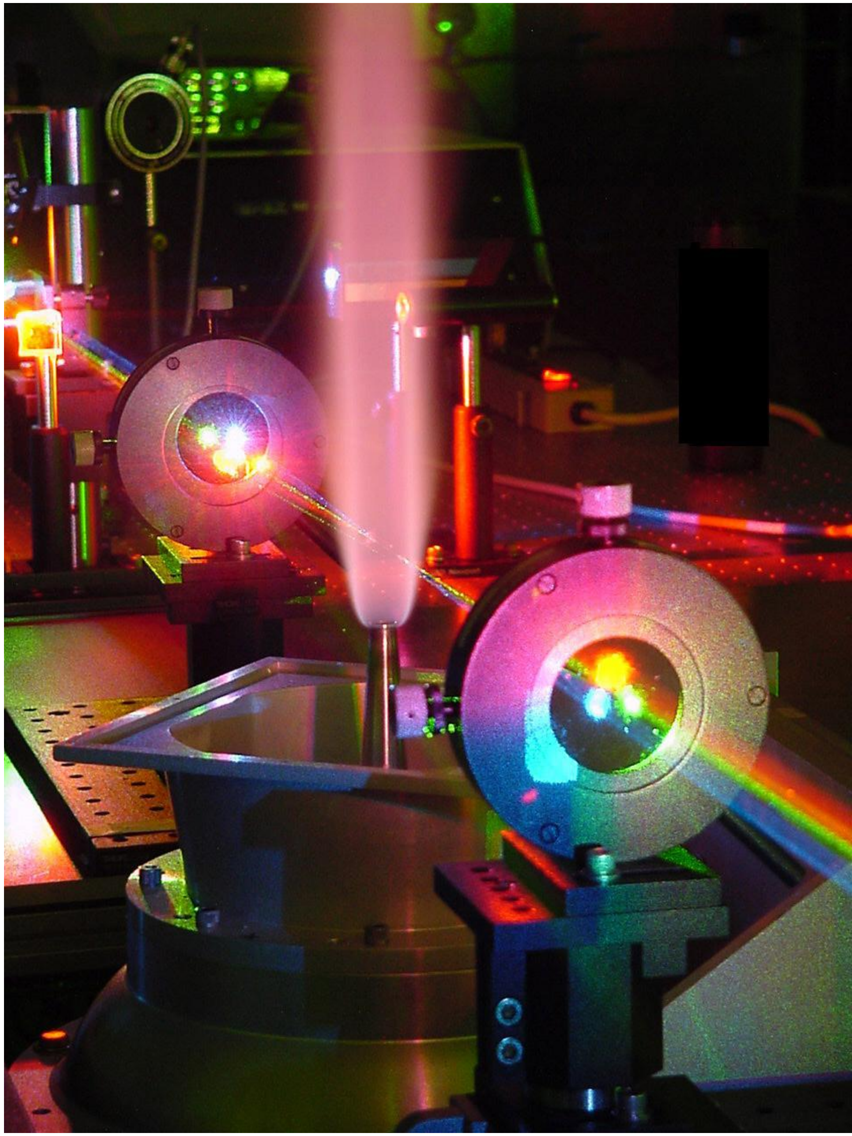
Ns CARS for Gas-Phase Diagnostics

- **Nonintrusive**
- **Coherent Laser-Like Signal**
- **Spatially and Temporally Resolved**
- **Excellent Gas-Temperature Measurement Technique**
- **Nonresonant Background**
- **Collisional/Linewidth Effects**
- **Data-Acquisition Rates: No Correlation Between Laser Shots at 10 Hz**
- **Broadband Dye Laser Mode Noise**



Good Bad Both

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Fsec CARS for Gas-Phase Diagnostics

- Fs lasers have much higher repetition rates than ns Q-switched Nd:YAG lasers: > 1 kHz versus ~10 Hz, plus can eliminate many of the other drawbacks – no need to measure collisional linewidths
- For application as a diagnostic probe for turbulent flames, signal levels must be high enough to extract data on a single laser shot from a probe volume with maximum dimension ~ 1mm.
- How effectively can Raman transitions with line width ~ 0.1 cm^{-1} line width be excited by the fs pump and Stokes beams (200 cm^{-1} bandwidth)? Answer: <u>very</u> effectively.
- How do we extract temperature and concentration from the measured single-shot fs CARS signals?

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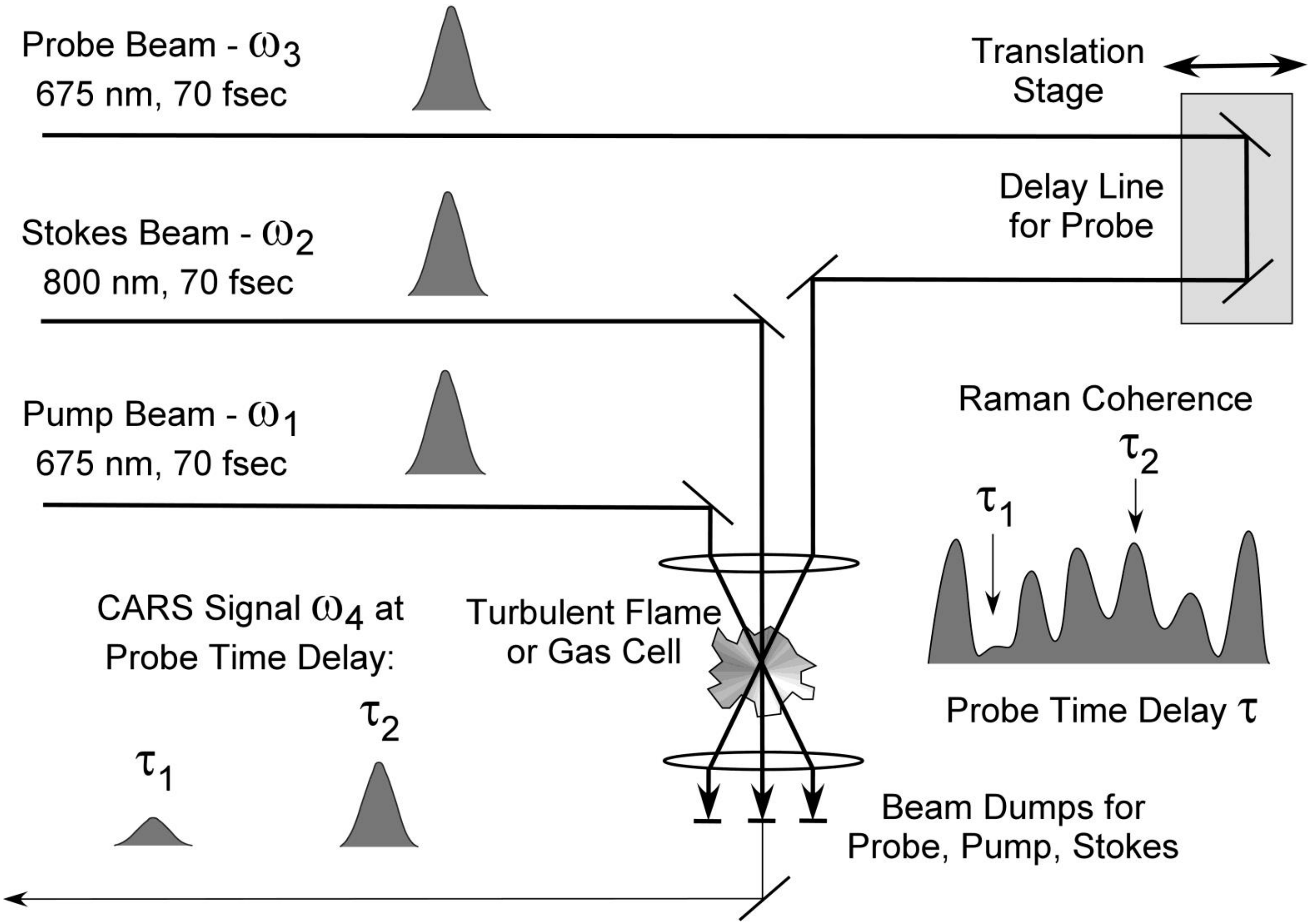
Optical System for Fs CARS with Mechanically Scanned Probe



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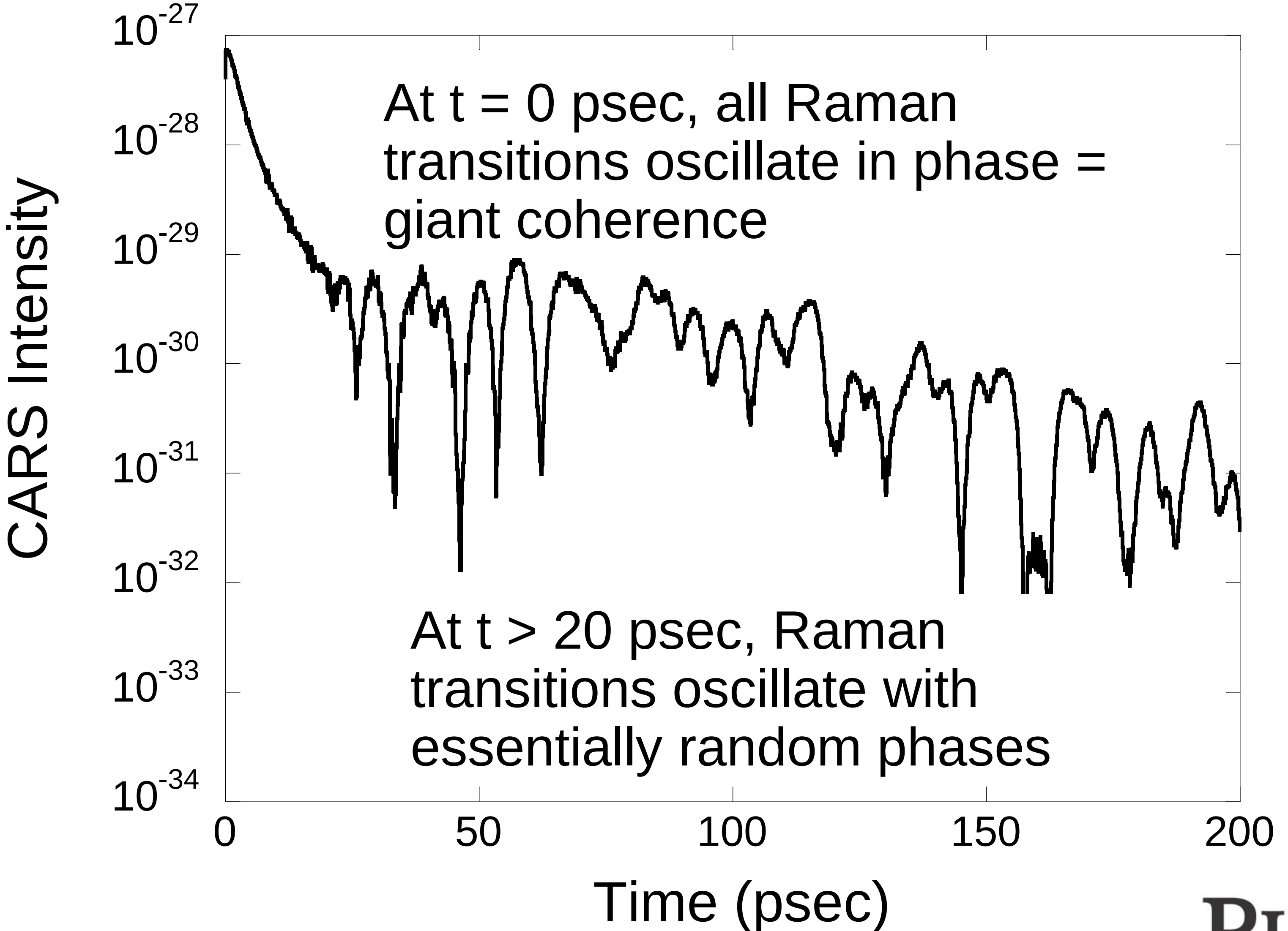
Calculated Time Dependence of CARS Intensity with Time-Delayed Probe Beam



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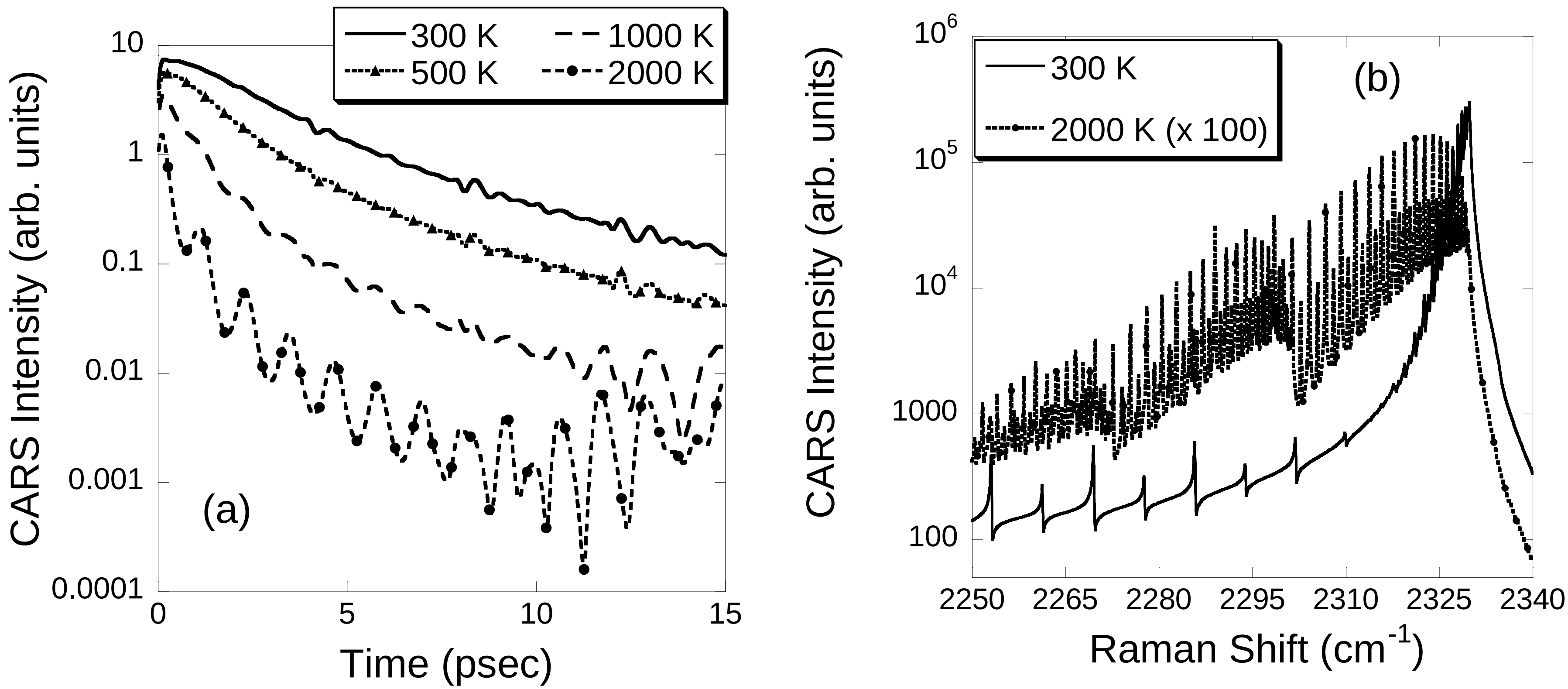

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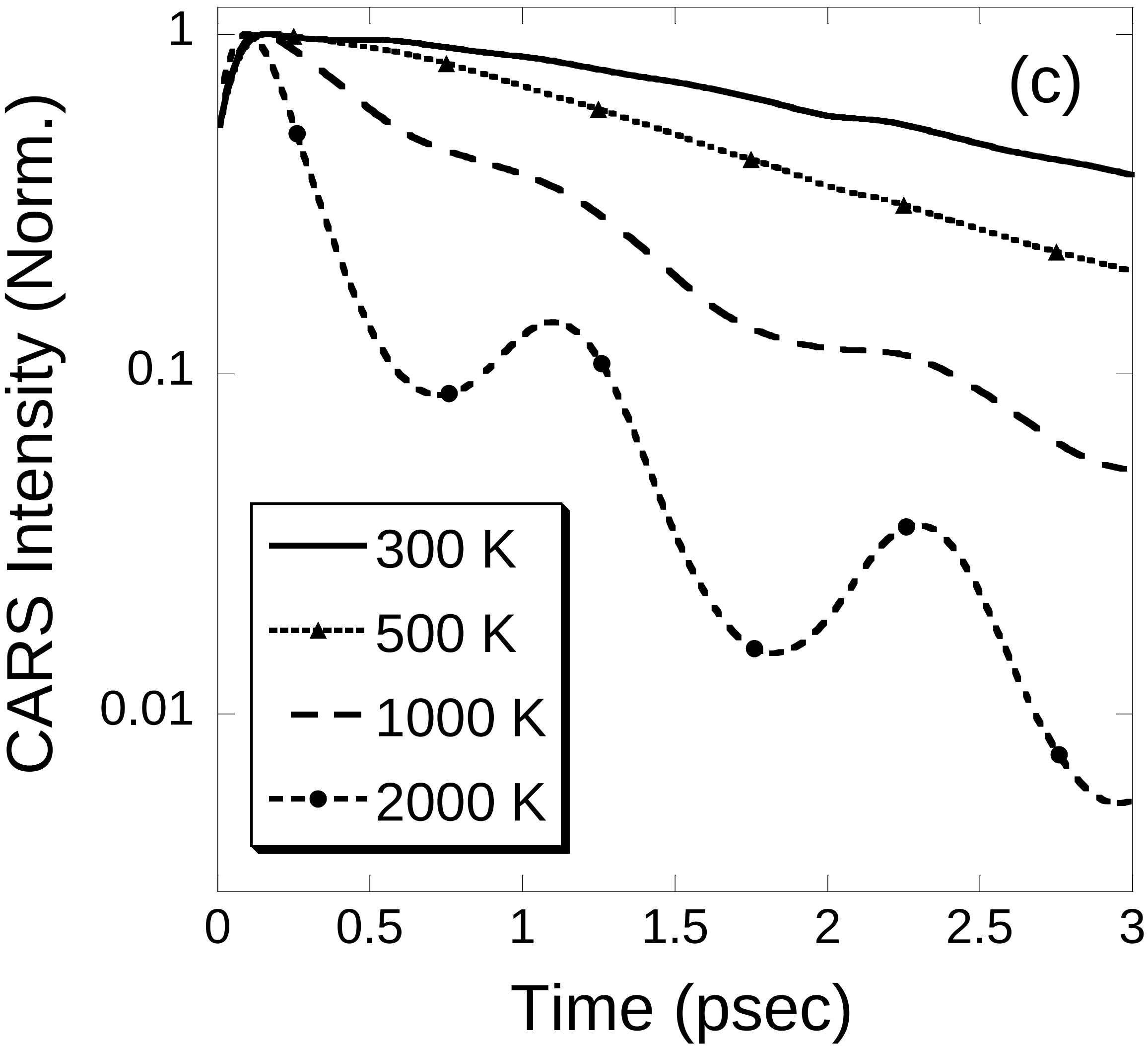

Temperature can be determined from the decay of the initial Raman coherence due to frequency-spread dephasing – Raman transitions oscillate with different frequencies.

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Raman Excitation for Fs Pump and Stokes Pulses

- There is a drastic difference in laser bandwidth (150-200 cm^{-1}) and Raman line width (0.05 cm^{-1}) for 100-fs pump and Stokes laser pulses.
- How effectively do the laser couple with the narrow Raman transitions?
- Can single-laser-shot fs CARS signals be obtained from flames? (The answer is yes)

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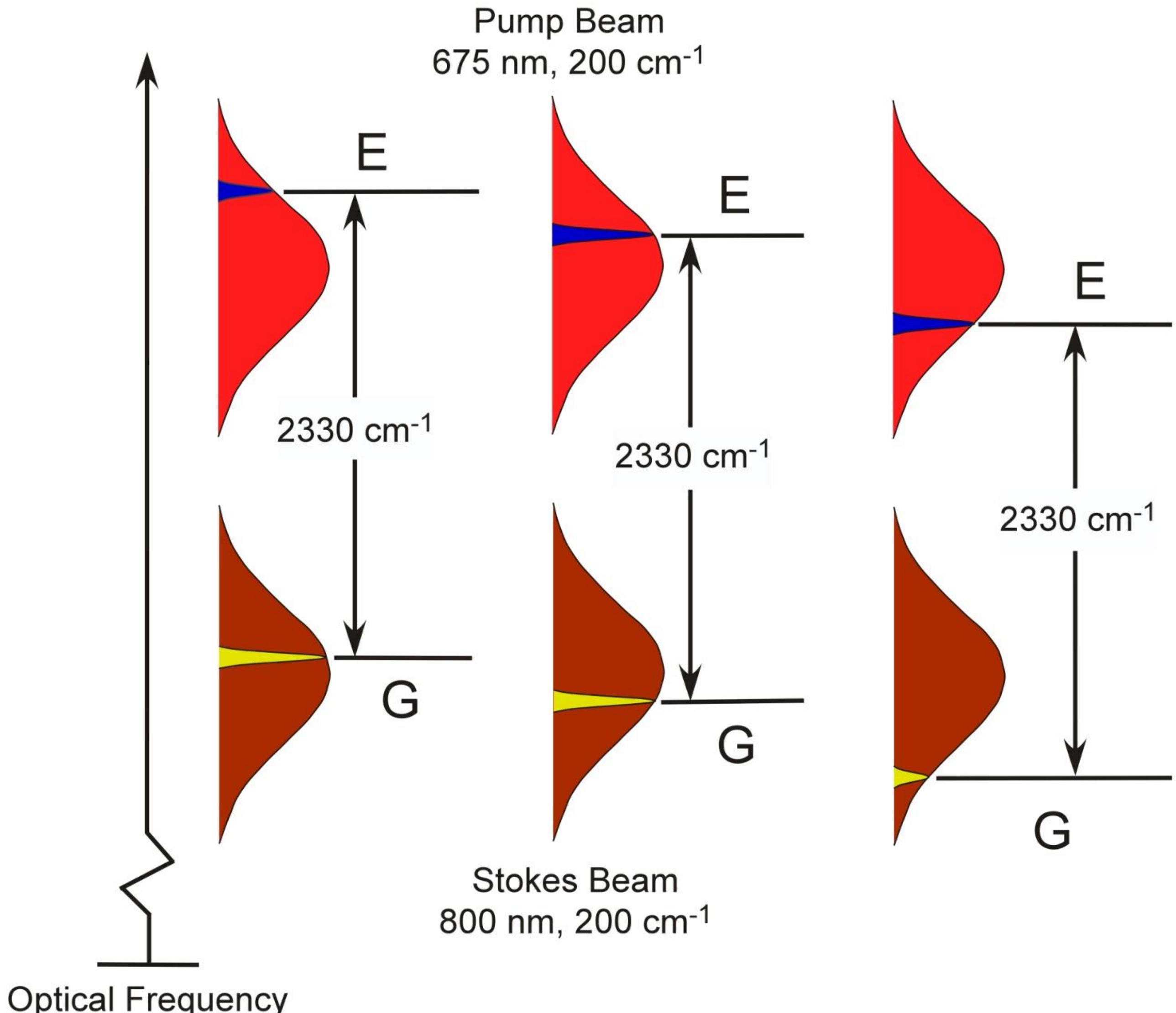
Coupling of 70-Fs Pump and Stokes Pulses with the Raman Coherence



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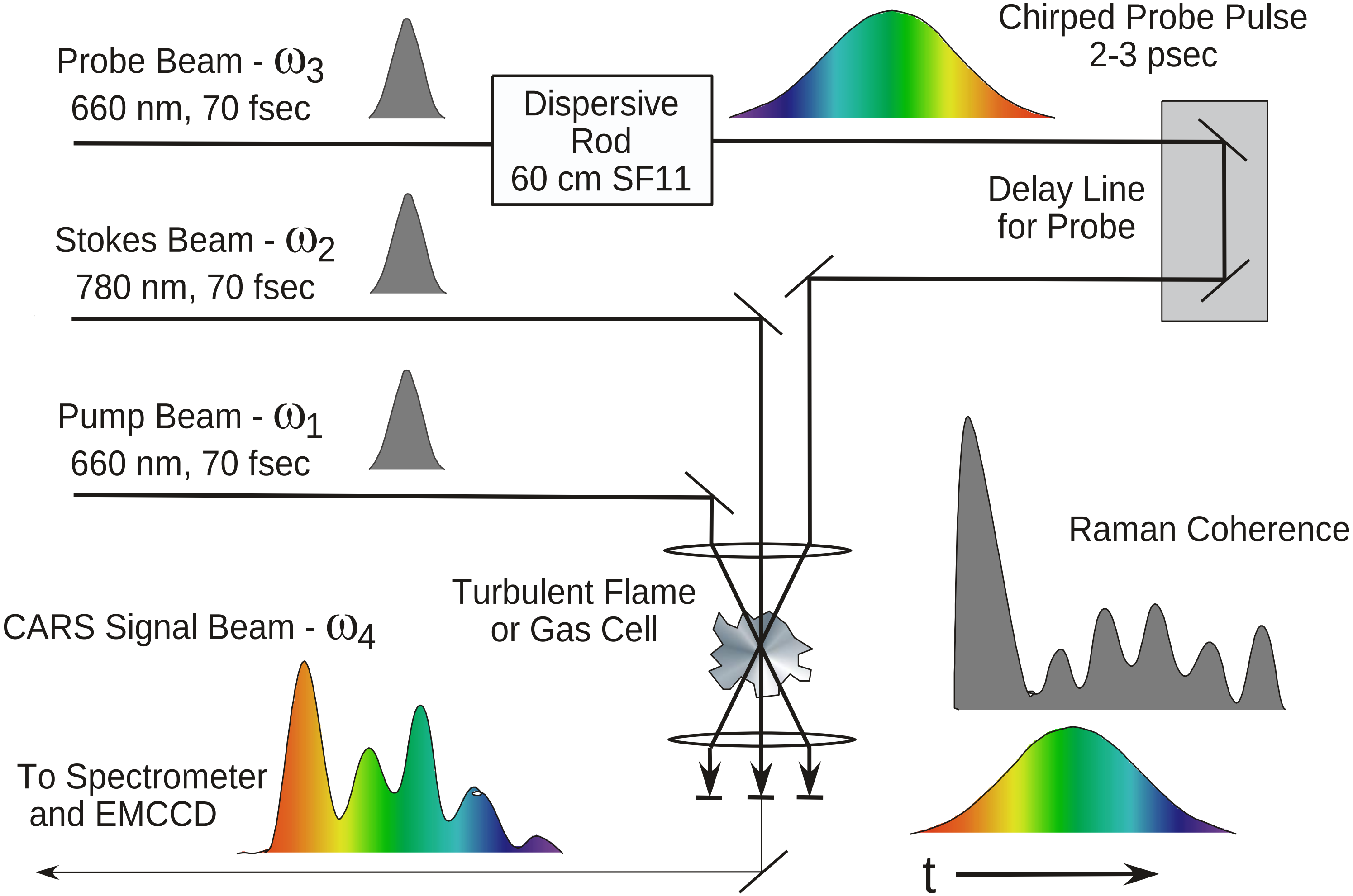
Optical System for Single-Pulse fs CARS with Chirped Probe Pulse



Lang and Motzkus, JOSA B **19**, 340-344 (2002).

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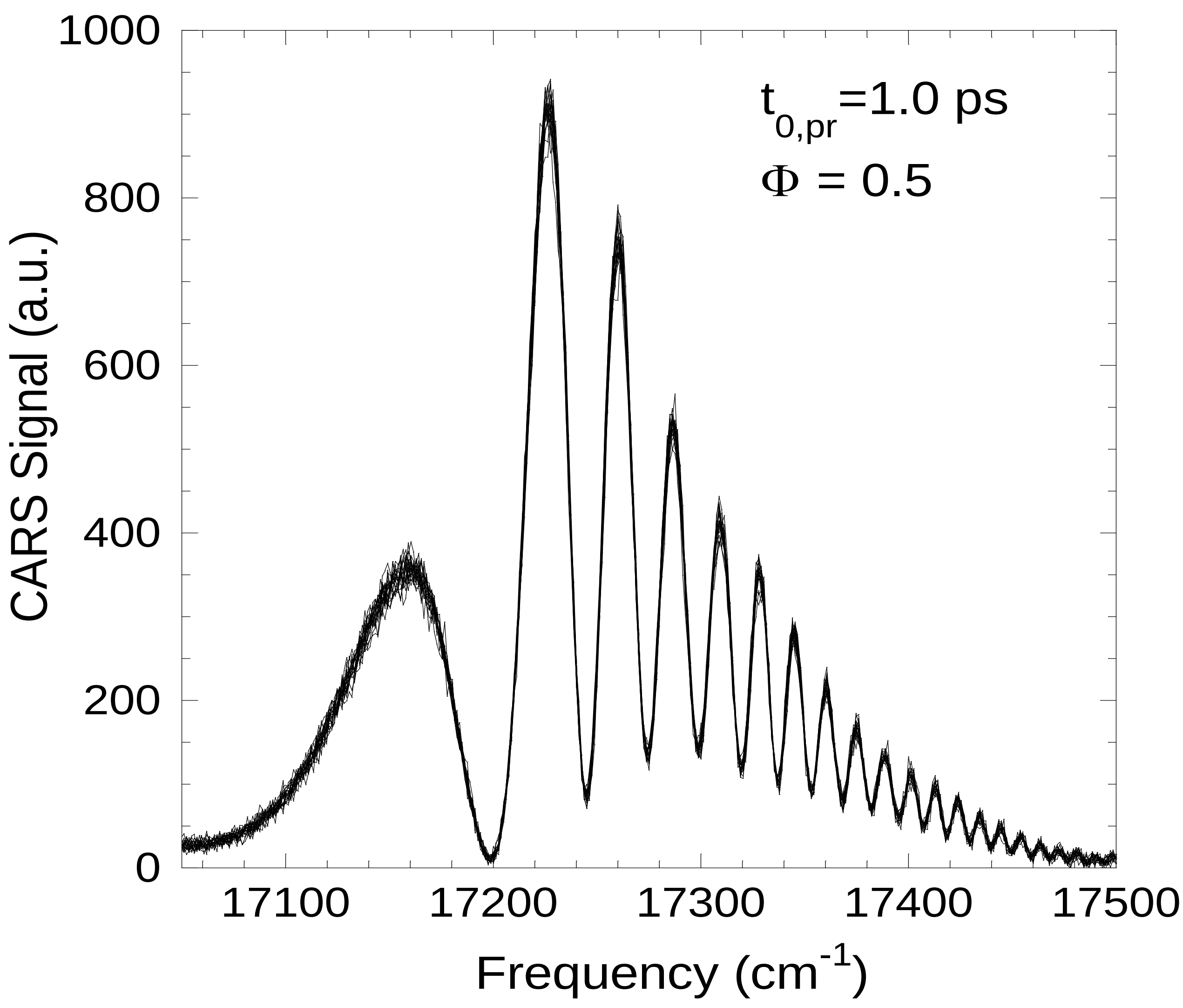
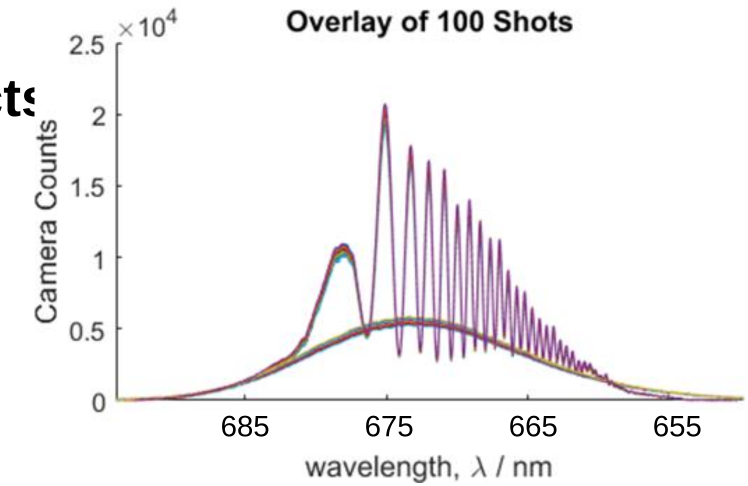
20 Single-Shots with Chirped Probe Pulse



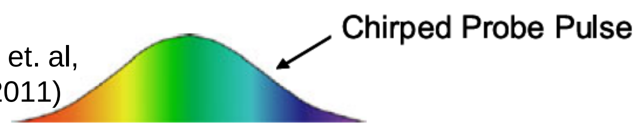
Chirped Probe Pulse fs CARS

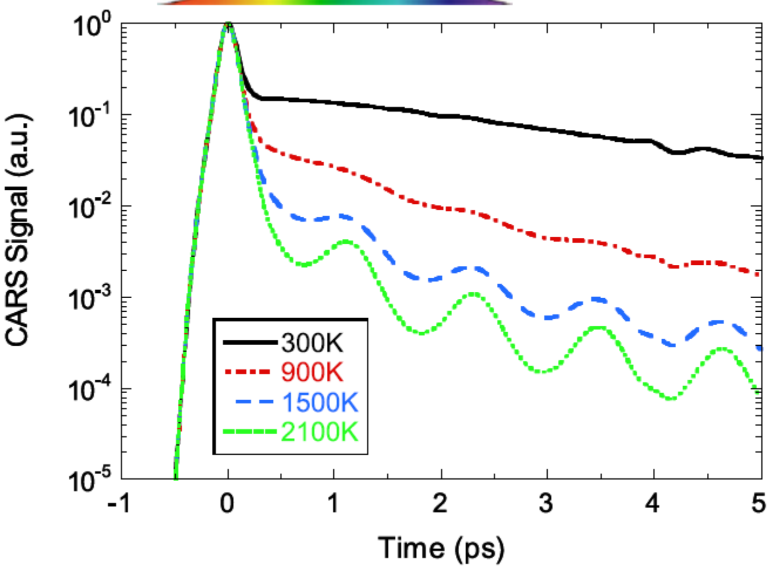
Ultrafast CARS

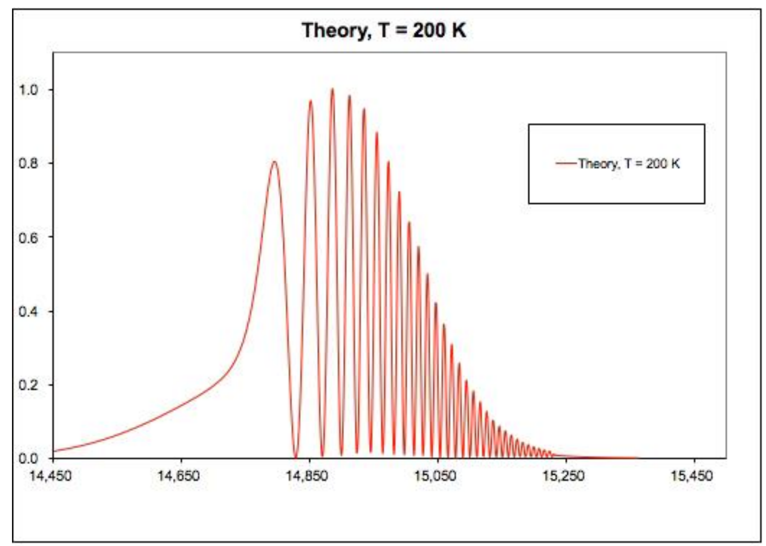
- Independent from collisional effects
- Stable, high PRF
- Good mode quality



ref: Richardson, et. al, Appl. Phys. B (2011)







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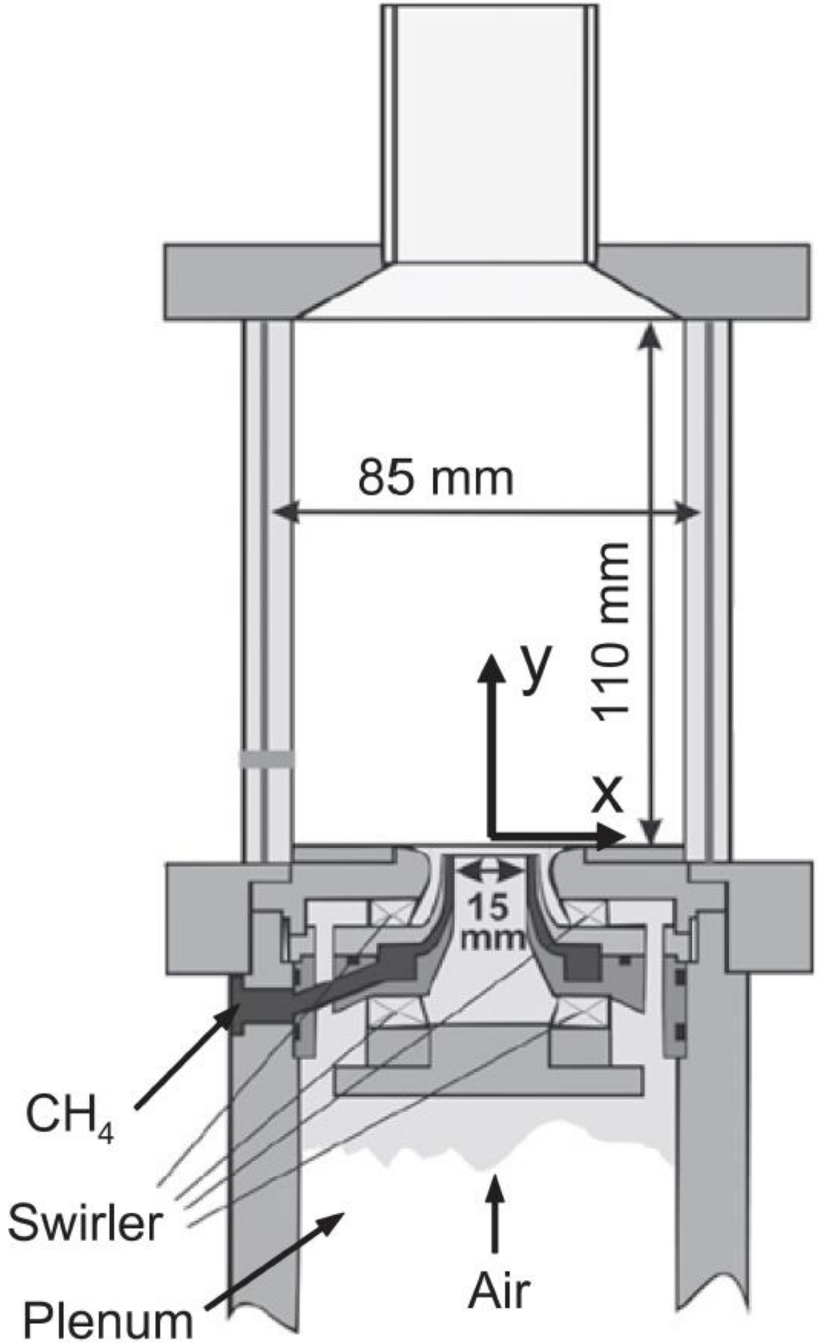
Single-Shot Temperature Measurements at 5 kHz: DLR Swirl-Stabilized Burner



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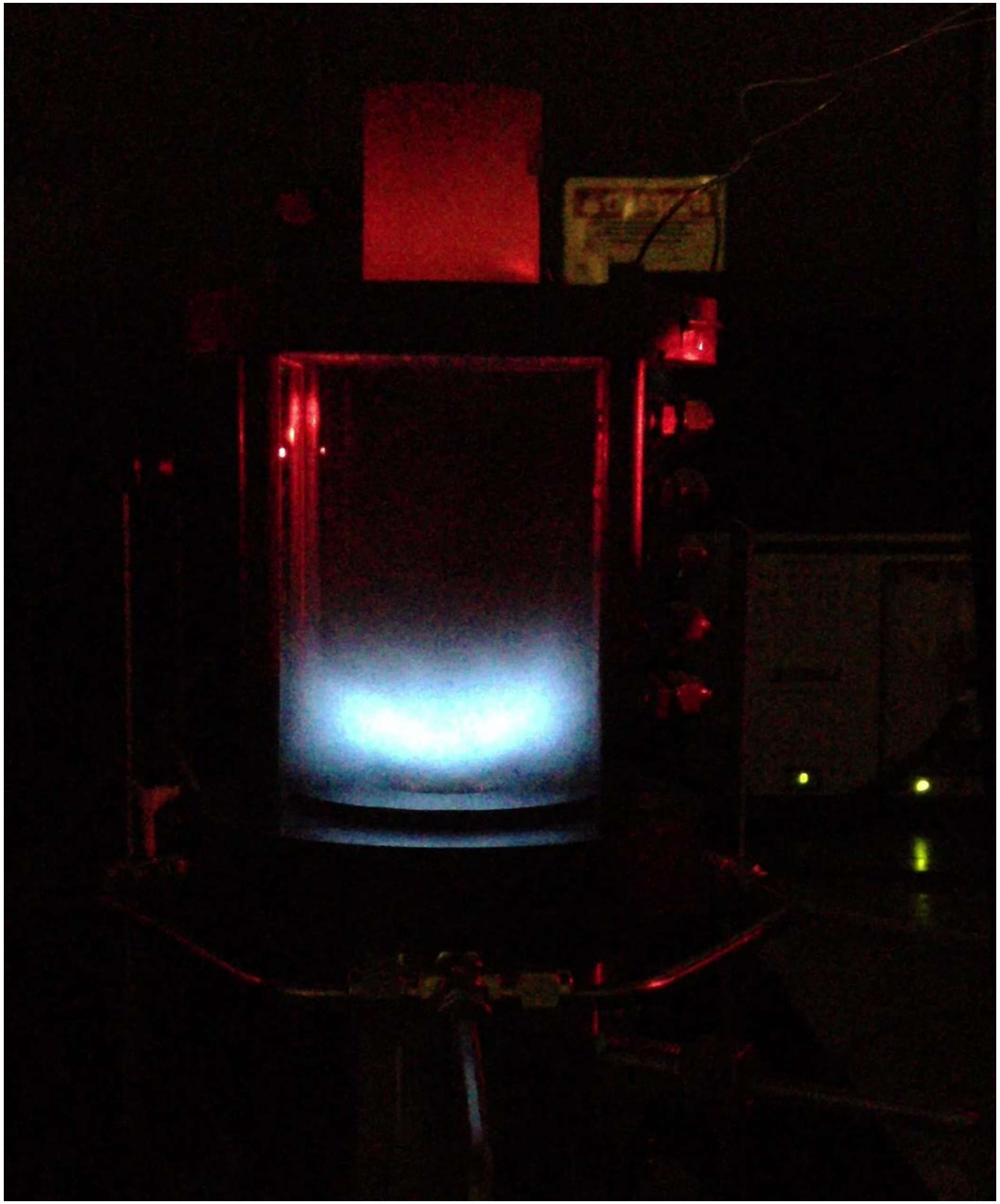

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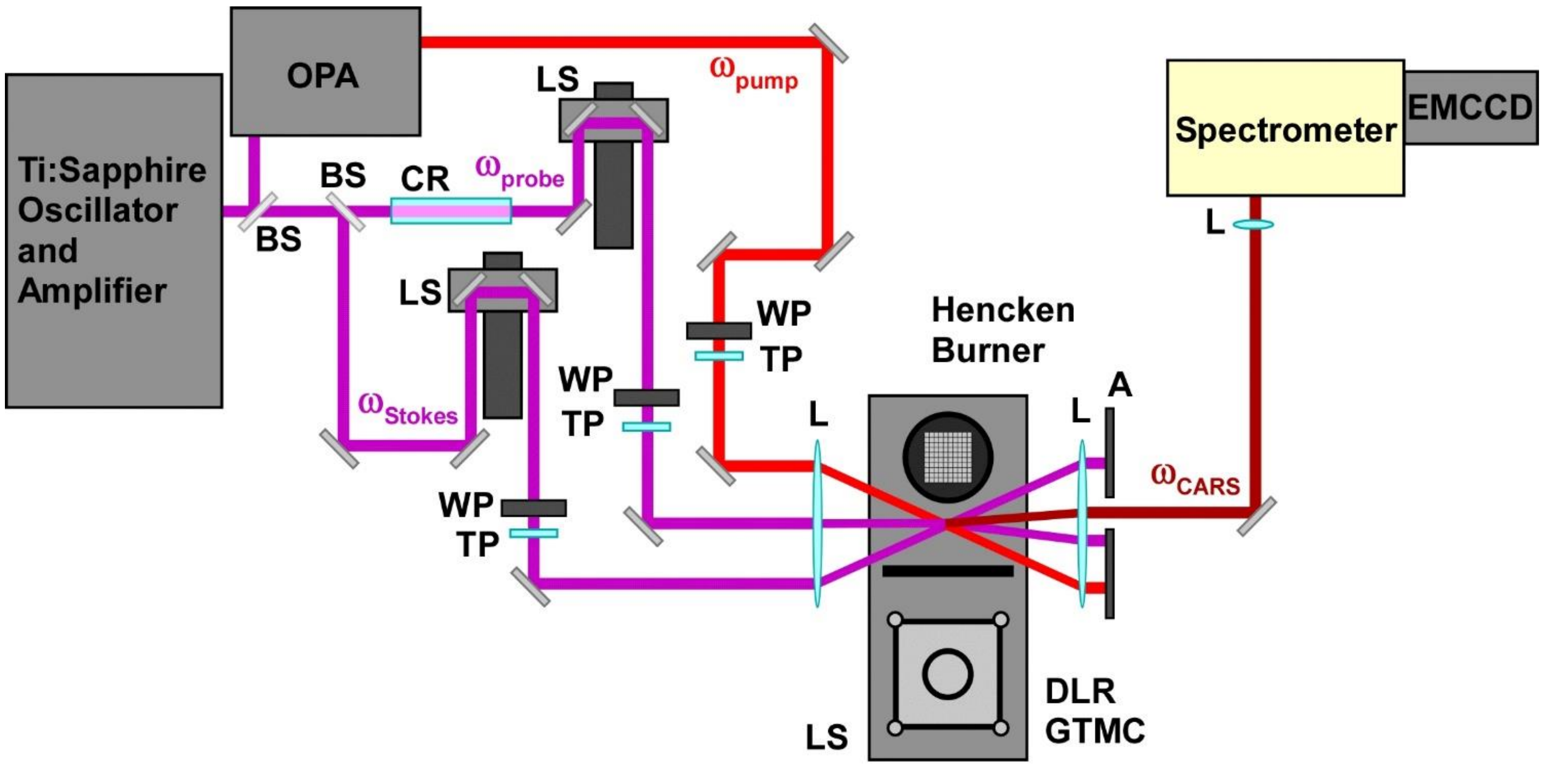

OPA: Optical Parametric Amplifier
WP: Zero Order Wave Plate
TP: Thin Film Polarizer
EMCCD: Electron Multiplying Charge-Coupled Device
BS: Beam Splitter
A: Aperture
LS: Linear Translation Stage
CR: Chirping Rod
L: Lens

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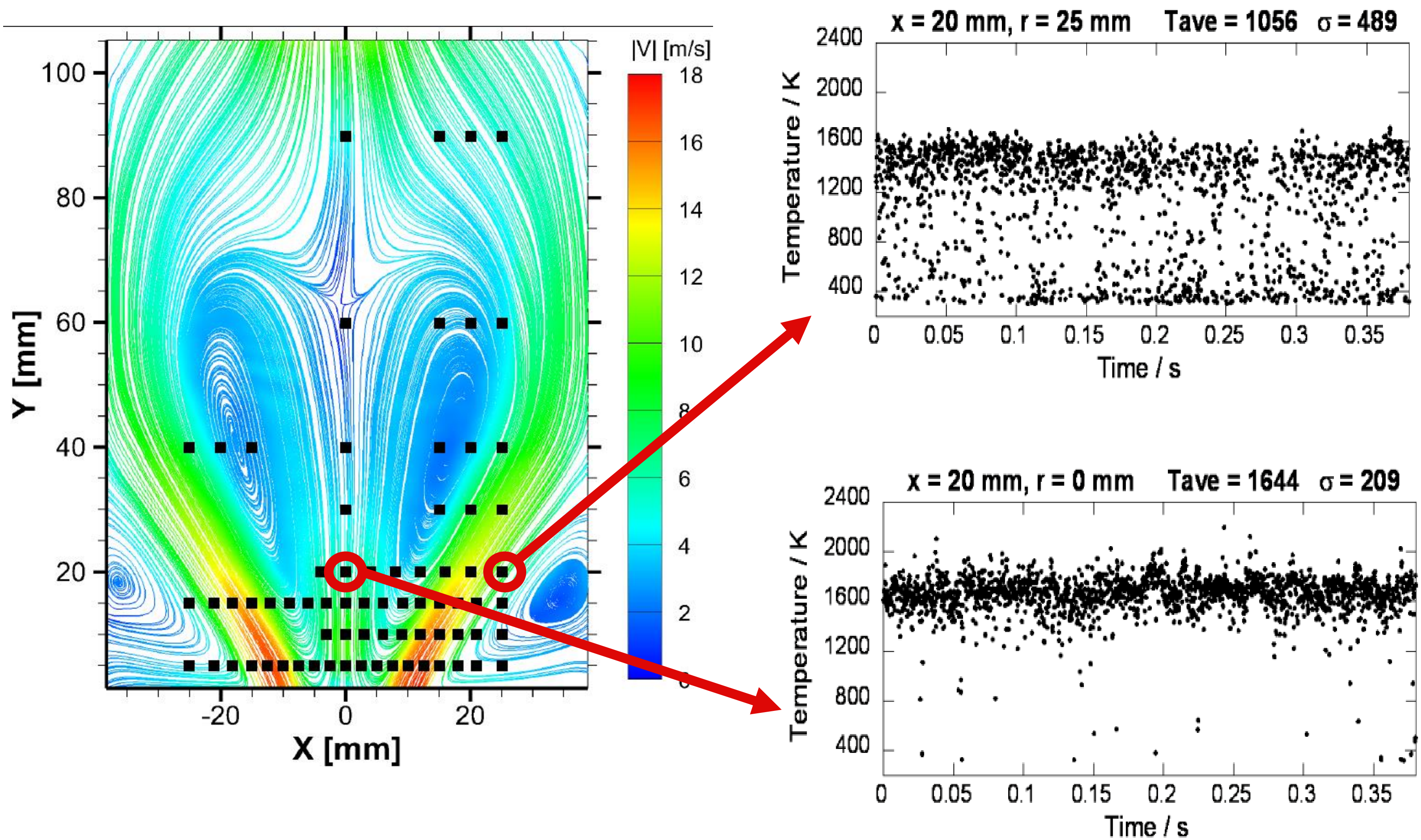
Single-Shot Temperature Measurements at 5 kHz: DLR Swirl-Stabilized Burner



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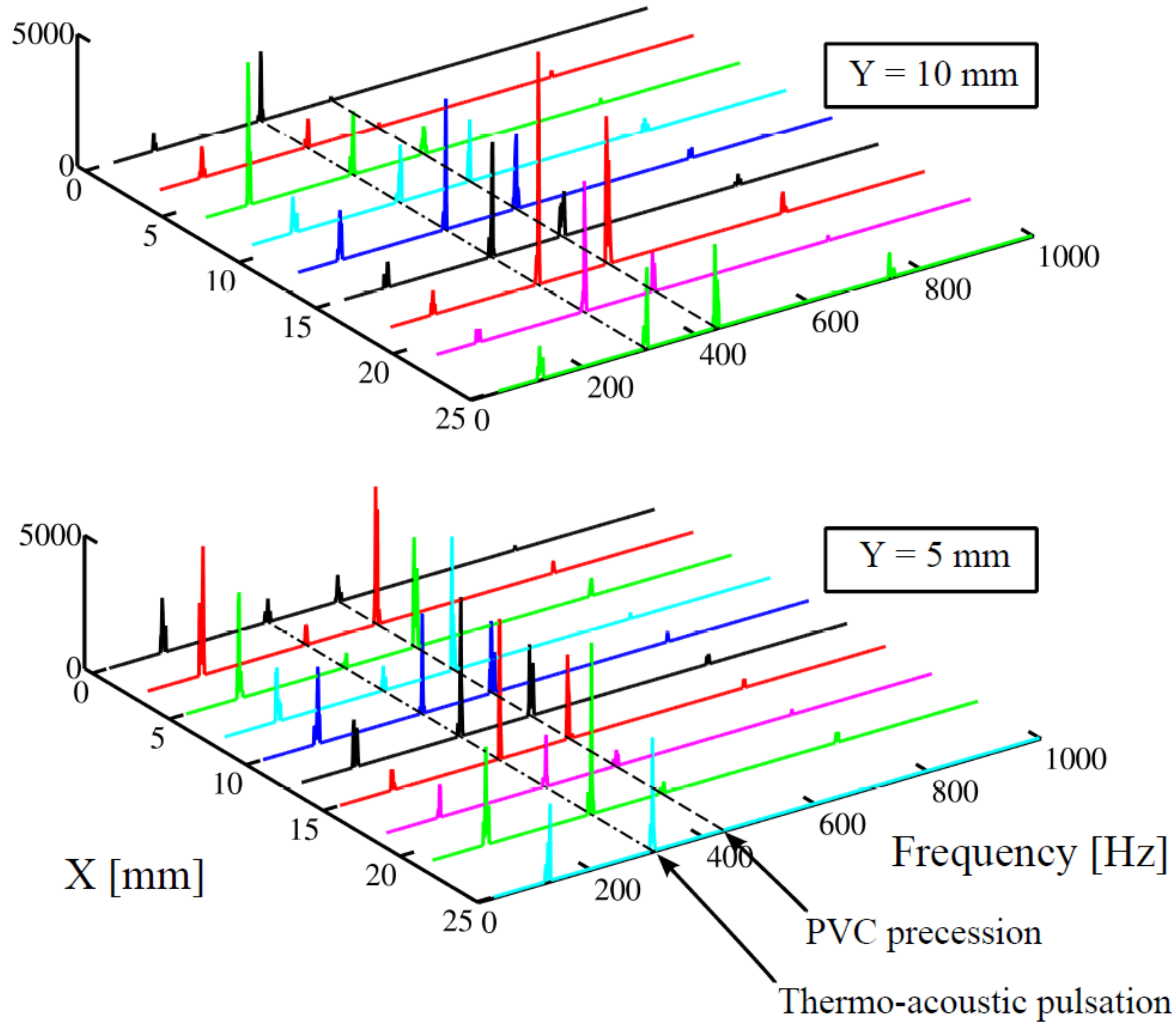
Single-Shot 5 kHz fs CARS Temp Measurements

Frequency conten of 5 kHz temperature measurements provides insight into the dynamics of large scale structures in the flow.



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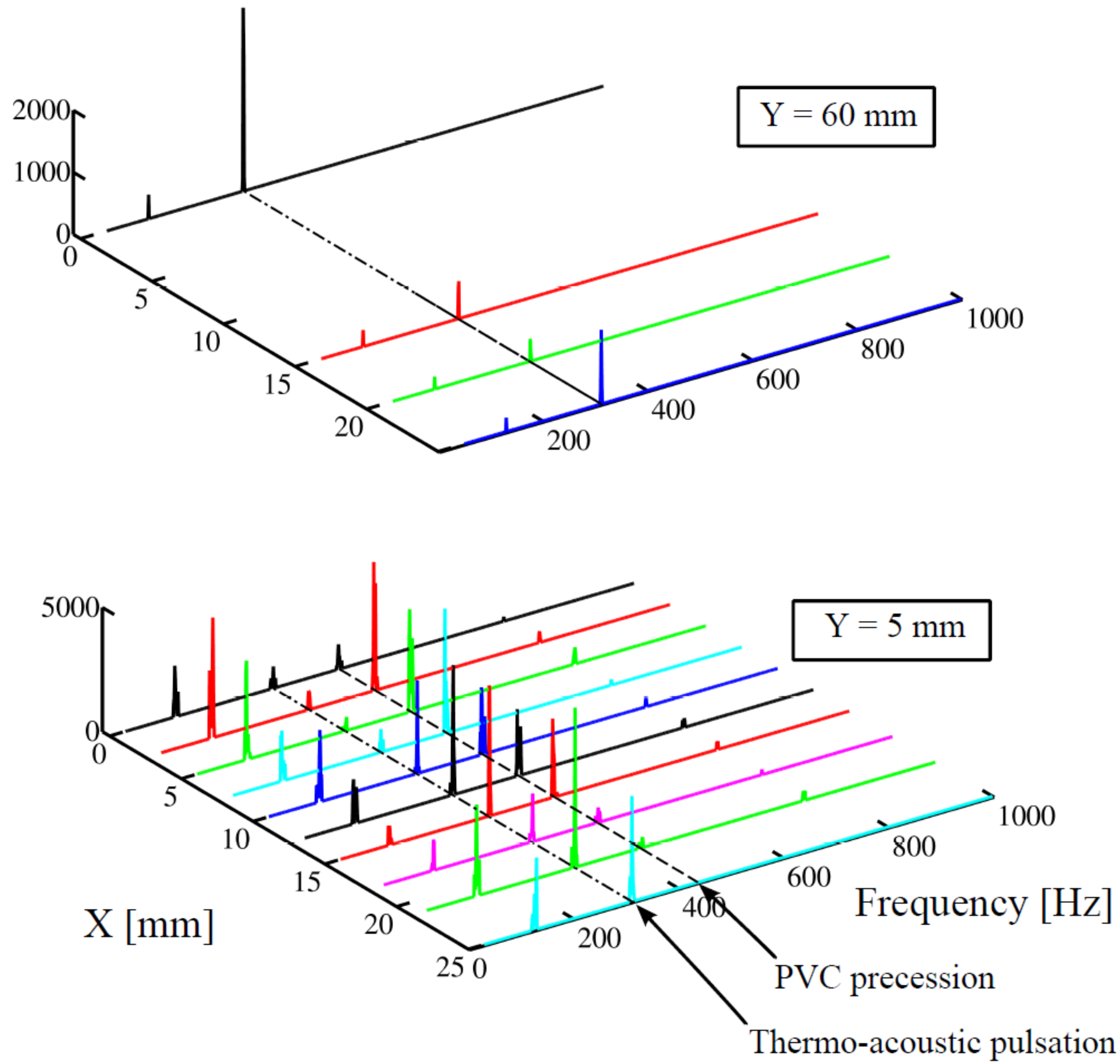

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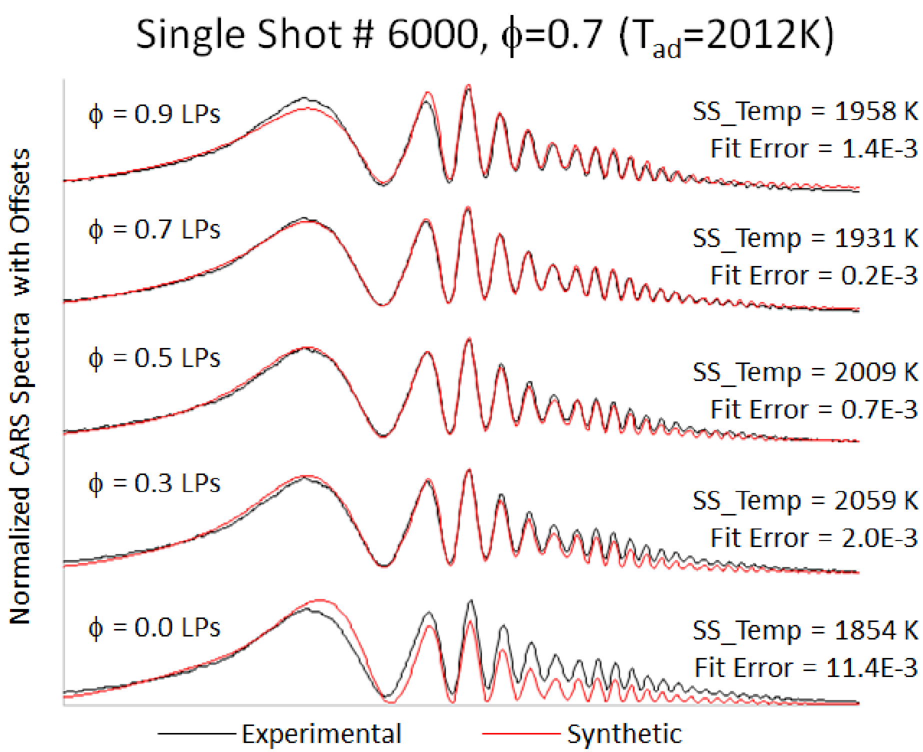
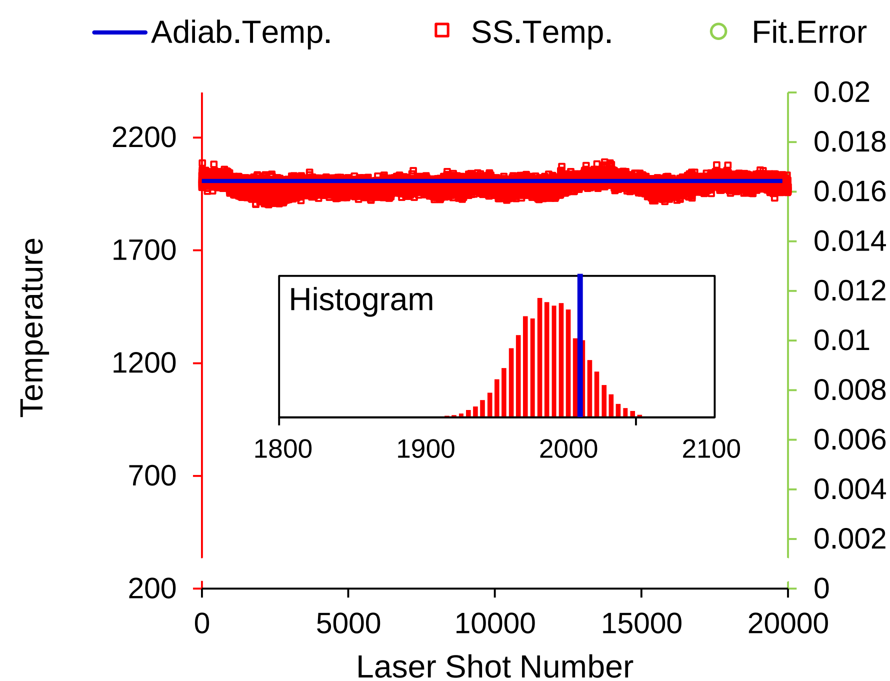
Spectral Fitting: Temperature Calibration





T_{avg} = 1989 K;
Accuracy = 1.17%;
Precision = ±1.19%

Single-Laser-Shot Temperature Measurement

- **Floating:**
  - **Temperature**
  - **Horizontal and vertical shift**
  - **Ratio of resonant to non-resonant response**

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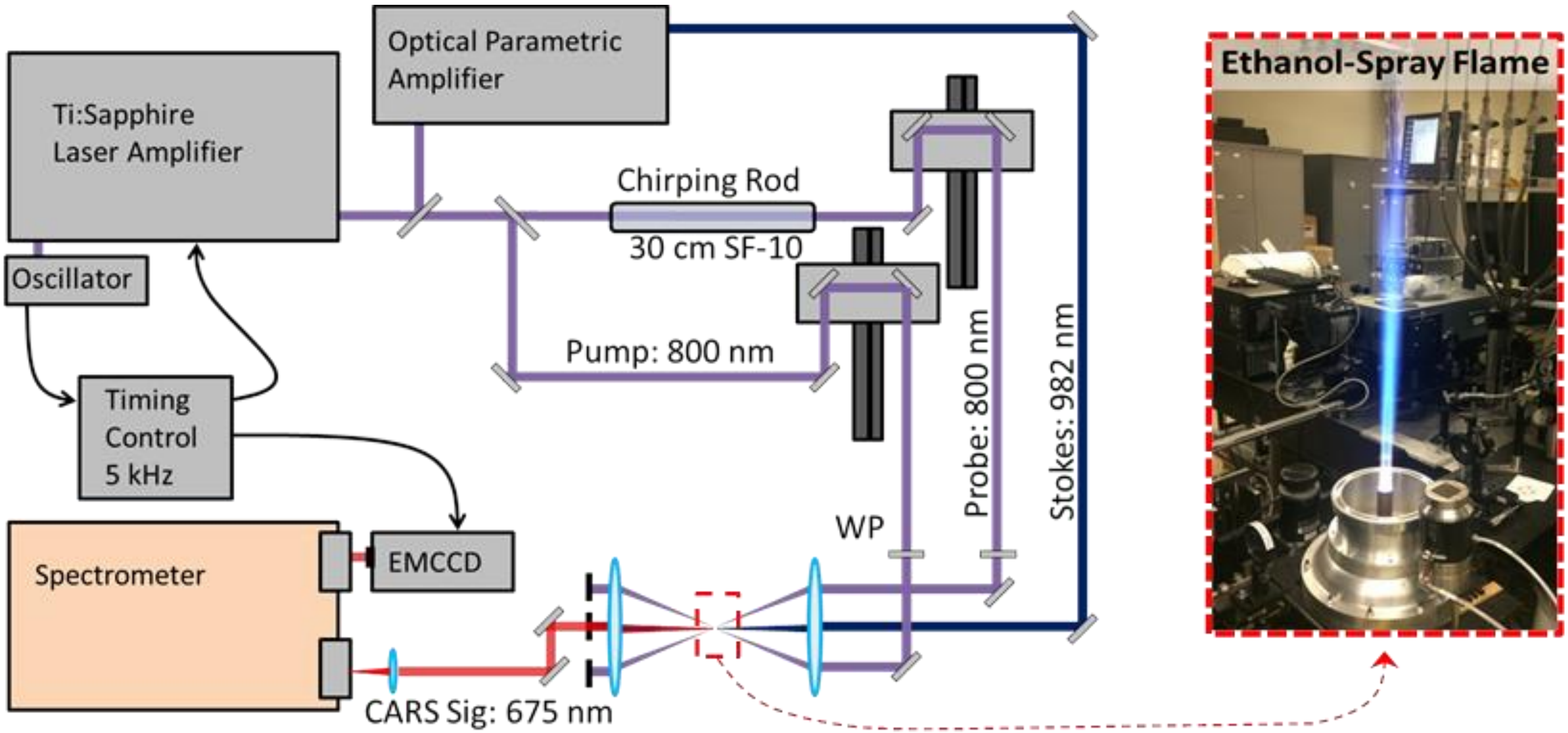
CPP fs-CARS LASER System – Overview



Probe Volume:

- Pump: 100 μJ, 60 fs, λ_o 800
- Stokes: 50 μJ, 60 fs, λ_o 982
- Probe: 200 μJ, 3.5 ps, λ_o 800
- Spatial resolution ~ 60 μm x 800 μm

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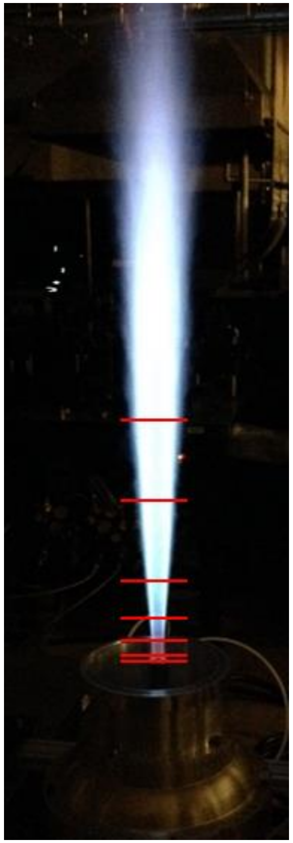
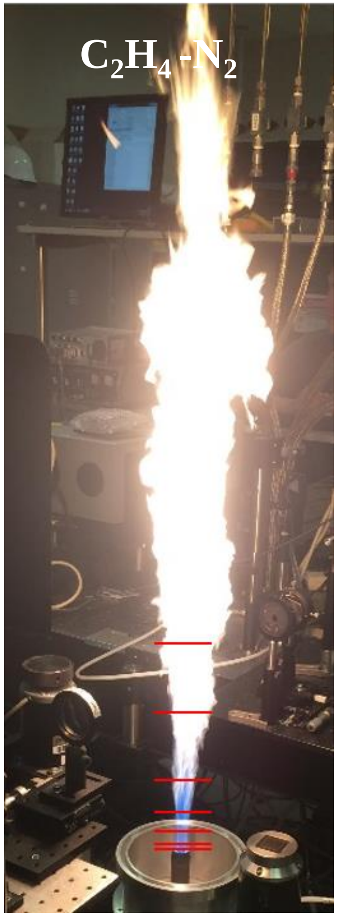
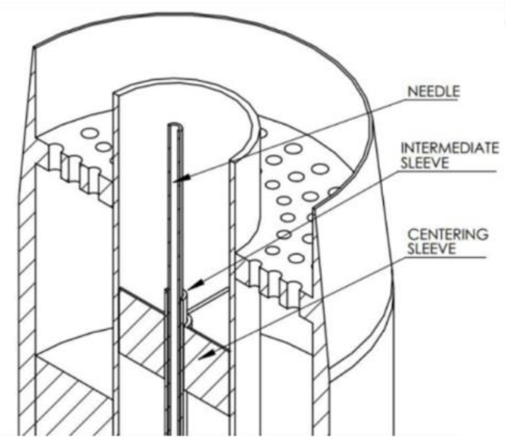
Univ. Sydney Needle Spray Burner (*SYNSBURN*[TM]) for Premixed, Diffusion, and Spray Flames

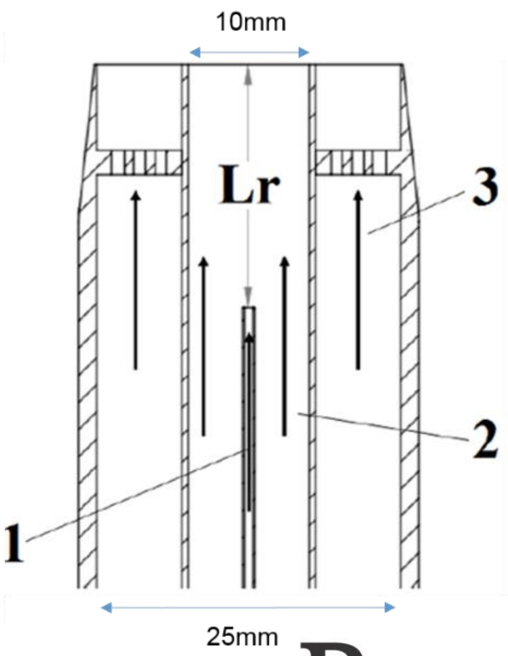
In addition to liquid-fuel spray flames:

- Premixed flame of methane-air
- Diffusion flame of ethylene-nitrogen









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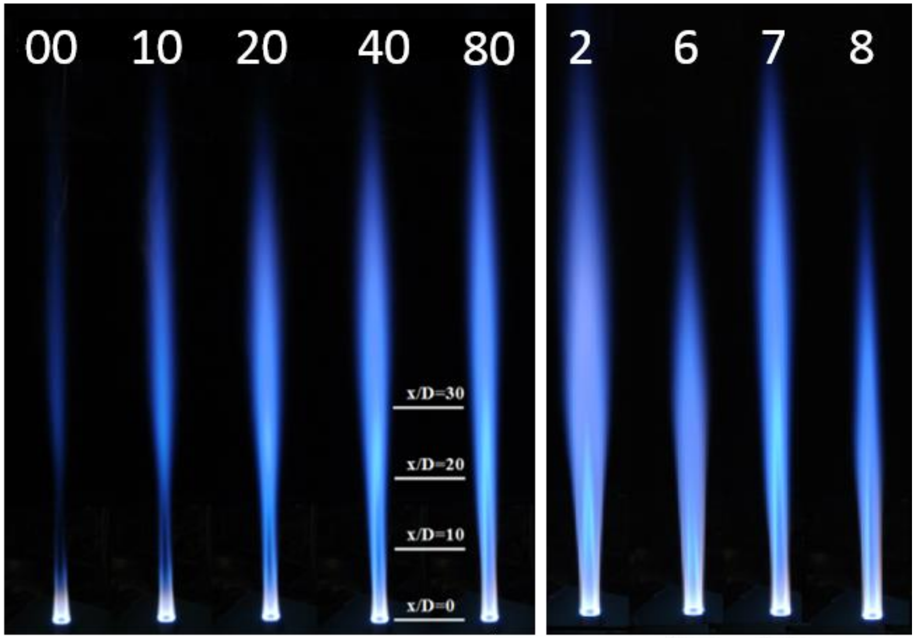
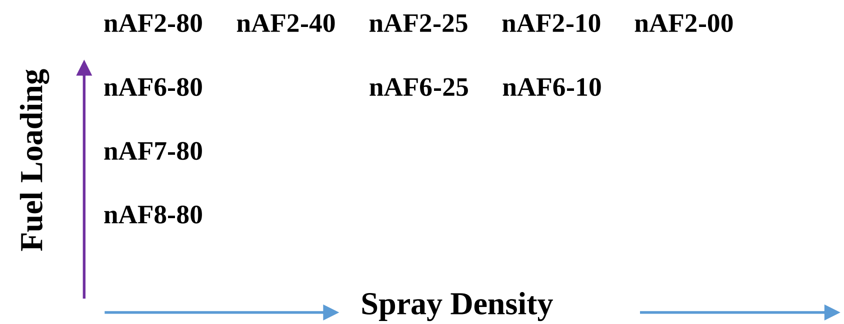
SYNSBURN^{TM} Offers Full Control Over Spray Flame Conditions



nAF7 - xx   nAFx-80

Acetone Flames Measured



Liquid Fuel Type

- Acetone, C_3H_6O
- Ethanol, C_2H_6O

	Fuel and Air Loadings				
Flame Case	nAF2	nAF6	nAF7	nAF8	
Liquid Fuel Flow	75	45	75	45	(g/min)
Airblast velocity	36	36	60	48	(m/s)
Air-to-Fuel ratio	0.36	0.22	0.22	0.16	(by mass)

Experimental Variables

- Fuel Loading
- Spray Density

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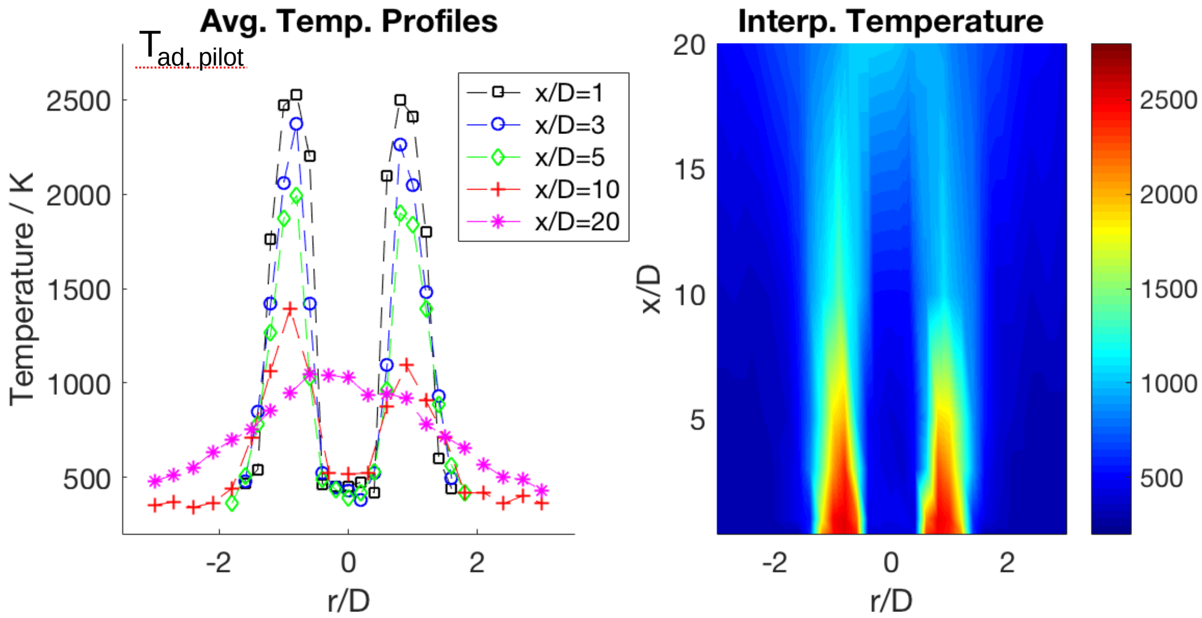
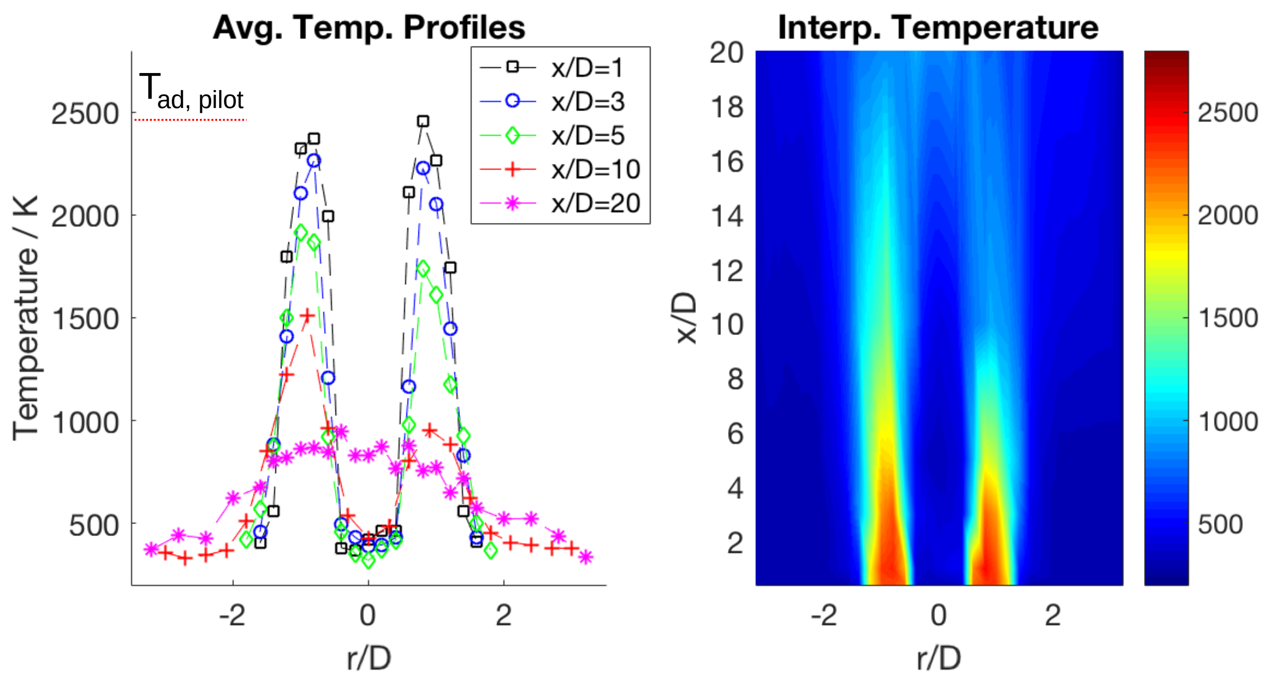
Hypothesis:
Heat release driven by droplet evaporation

Test:
Acetone significantly more volatile than ethanol

Observation:
Increased acetone temperature profile at x/D = 20



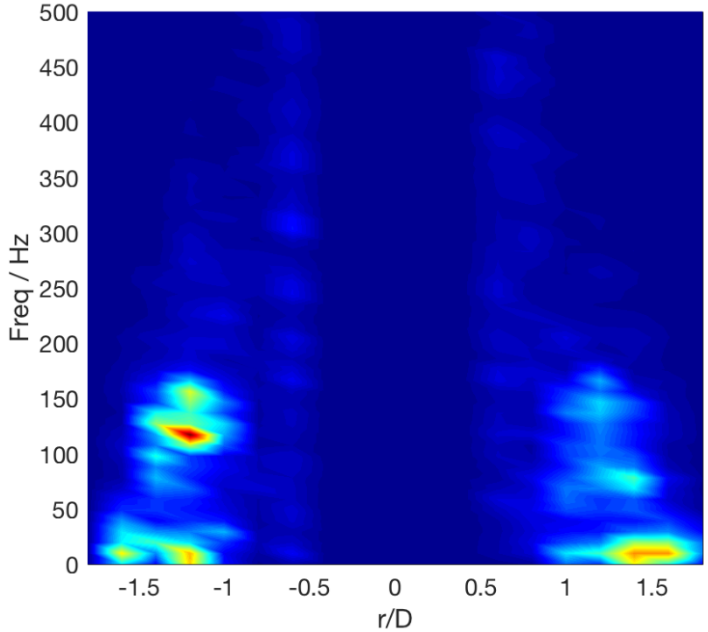
N-EF8-25 -- (C_2H_6O)

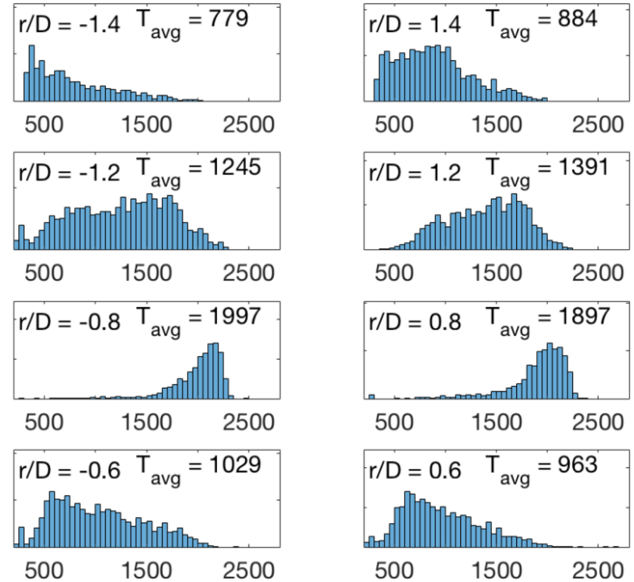


N-EF8-25 -- (C_2H_6O)

Temperature Spectrum at x/D = 5.0

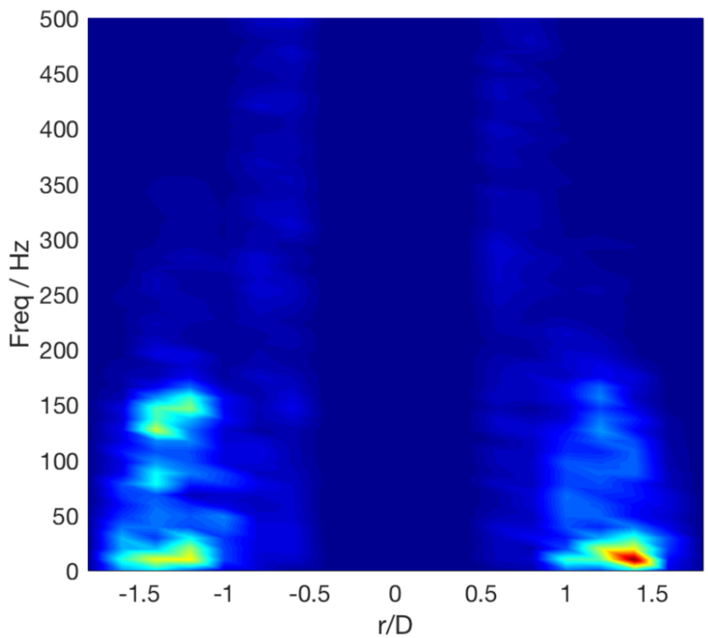
N-EF8-25 -- (C_2H_6O)

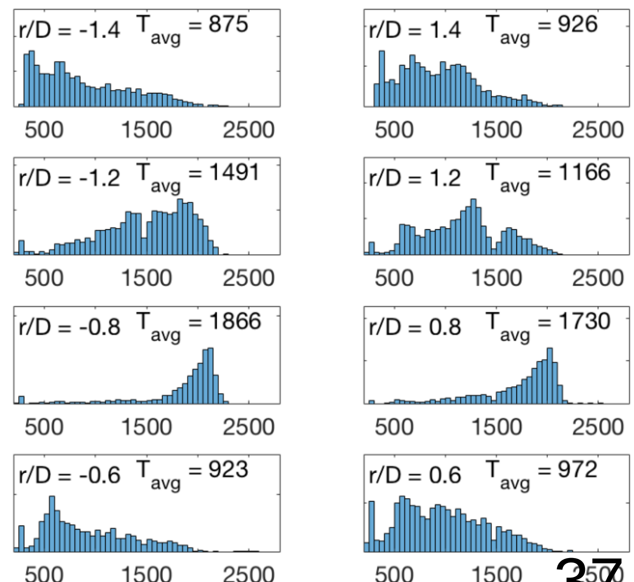




Tight distribution over pilot flame: r/D = 0.8

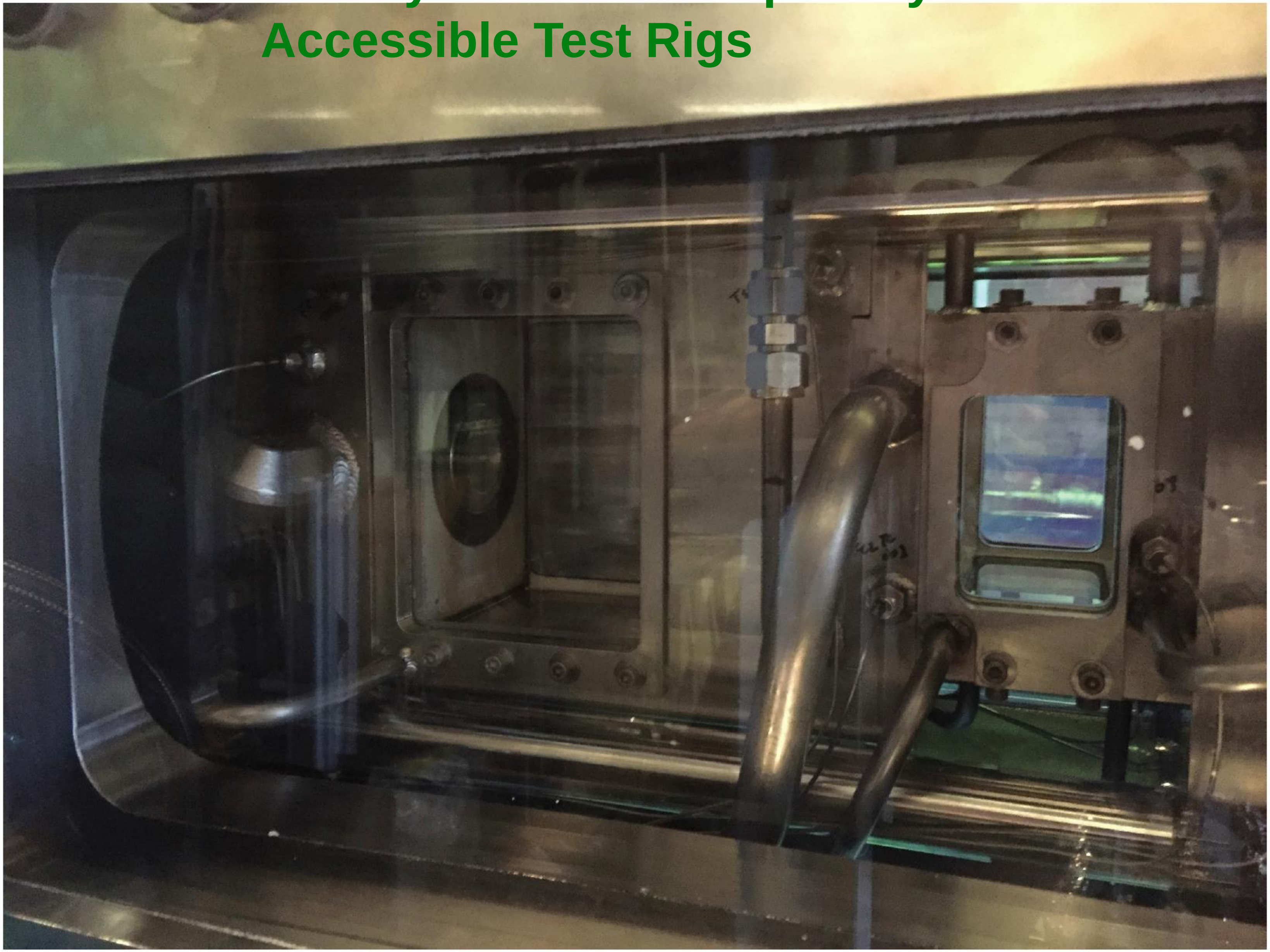
N-EF8-25 -- (C_2H_6O)



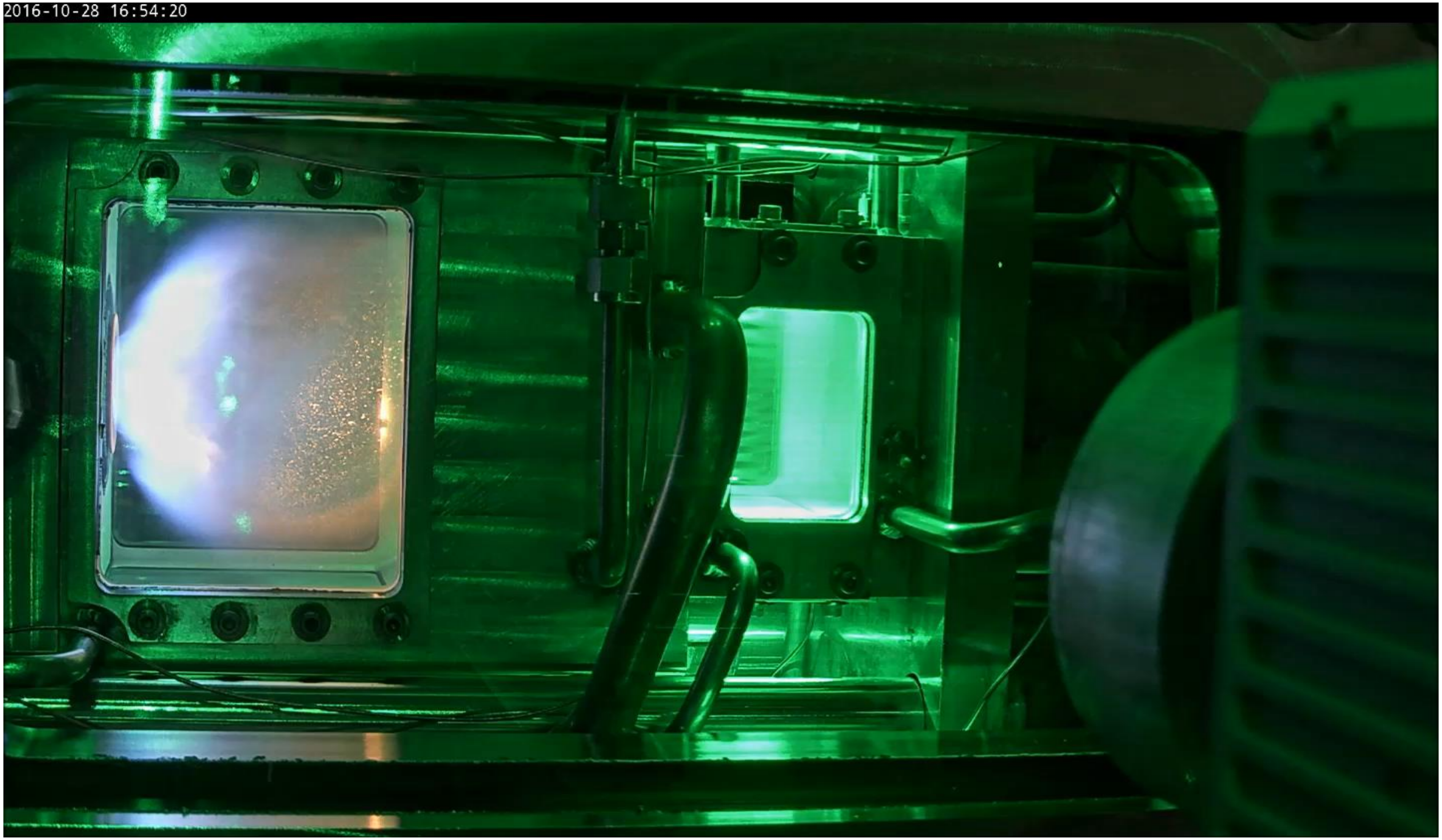


Wider spread at inner and outer pilot flame interface regions: r/D = 0.6 & r/D > 1.0

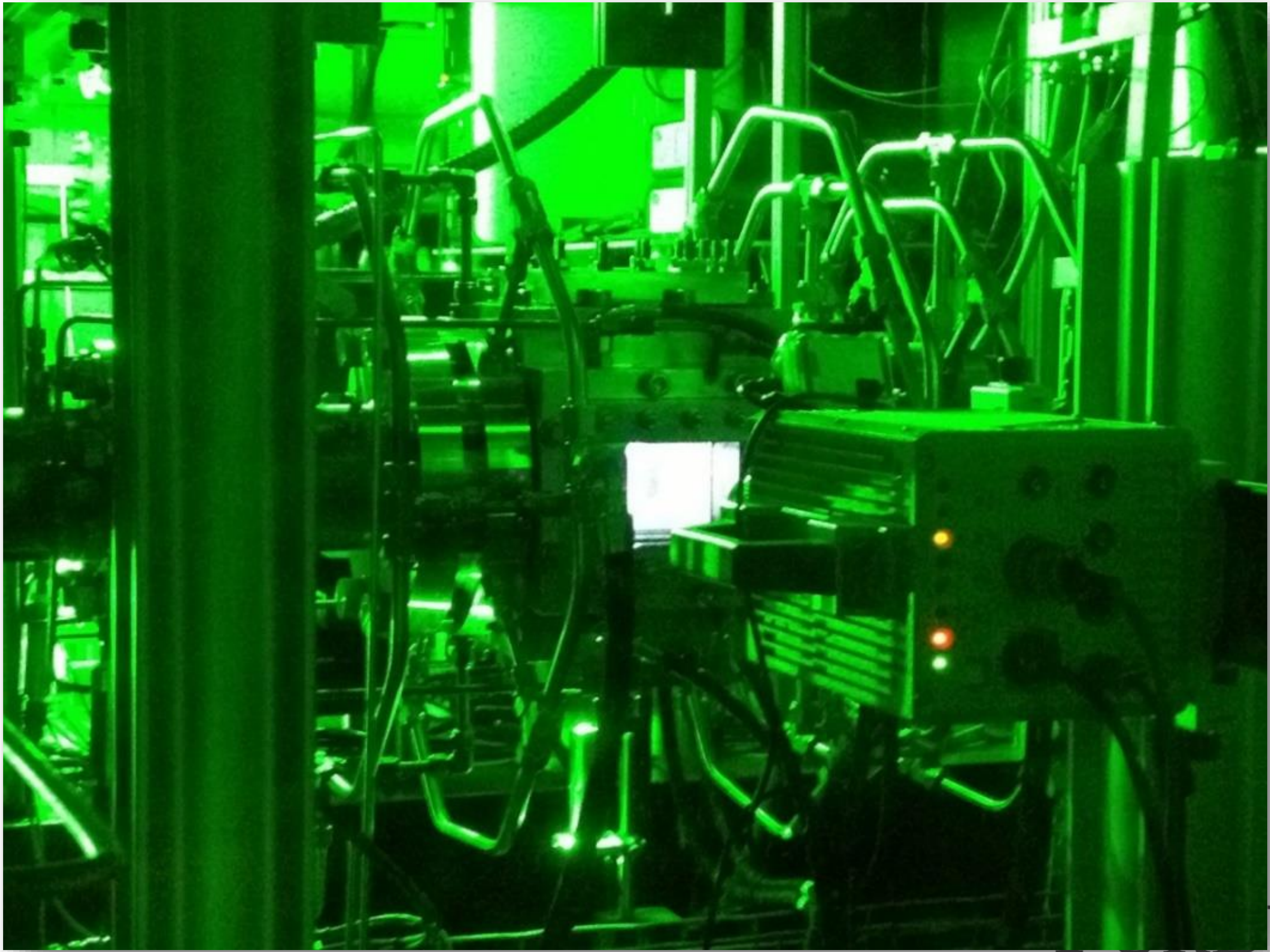
Purdue Gas Turbine Combustion Facility: Advanced Optically Accessible Test Rigs



Fs CARS: High-Pressure Aviation Gas Turbine Test Rig



Gas Turbine Combustion Test Rig: PIV



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Potential Advantages of fs CARS

- Data rate of 1-100 kHz would allow true time resolution, study of turbulent fluctuations and of transient combustion events
- Data rate of 1-100 kHz as opposed to 10 Hz would decrease test time considerably in practical applications
- Fs CARS, unlike ns CARS, is insensitive to collision rates even up to pressure > 10 bars (fs CARS signal increases with square of pressure)
- Fs laser pulses are near-Fourier-transform limited, noise may be decreased significantly for single-shot measurements

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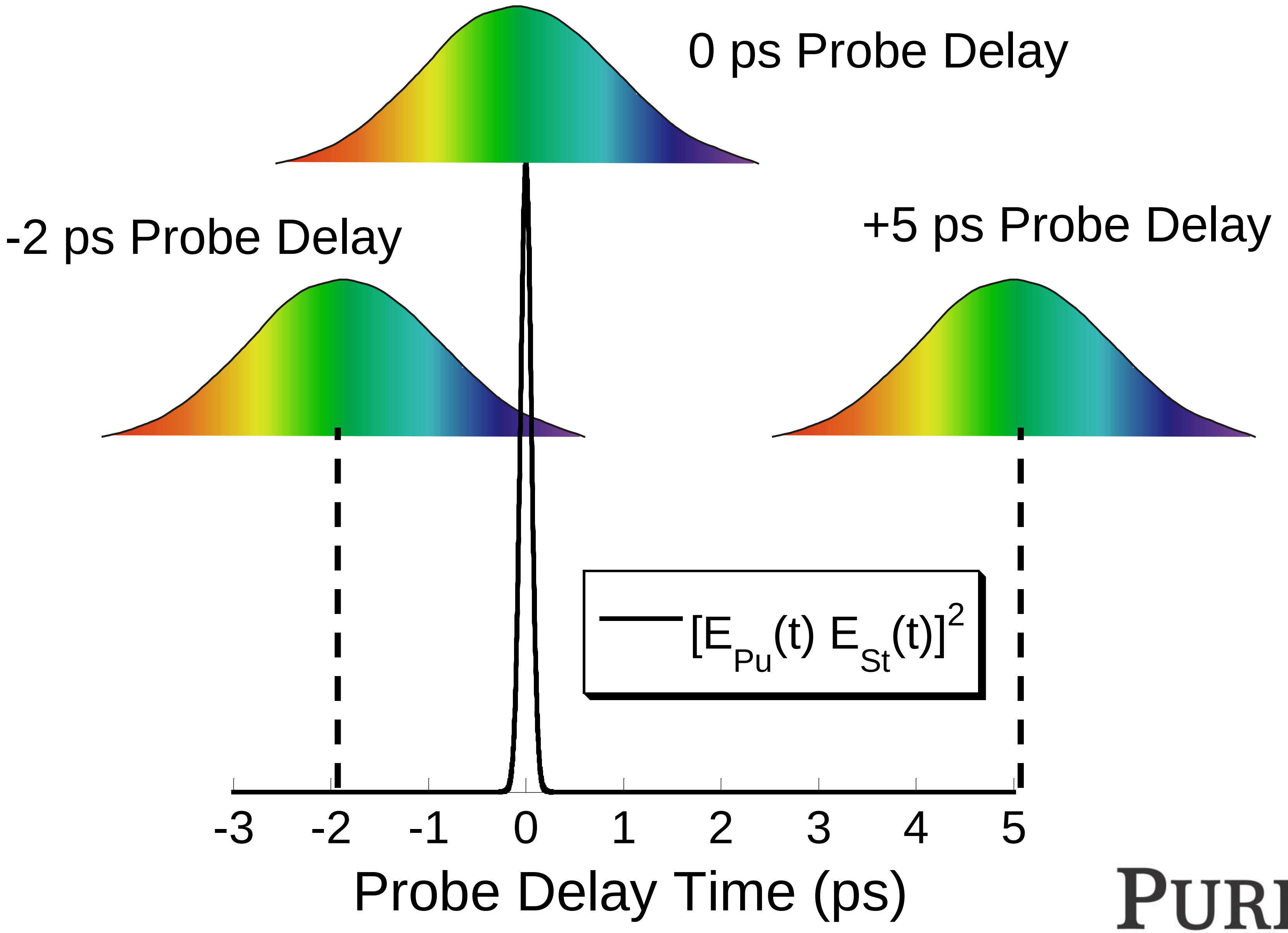
Chirped Probe Pulse (CPP) fs CARS in Argon



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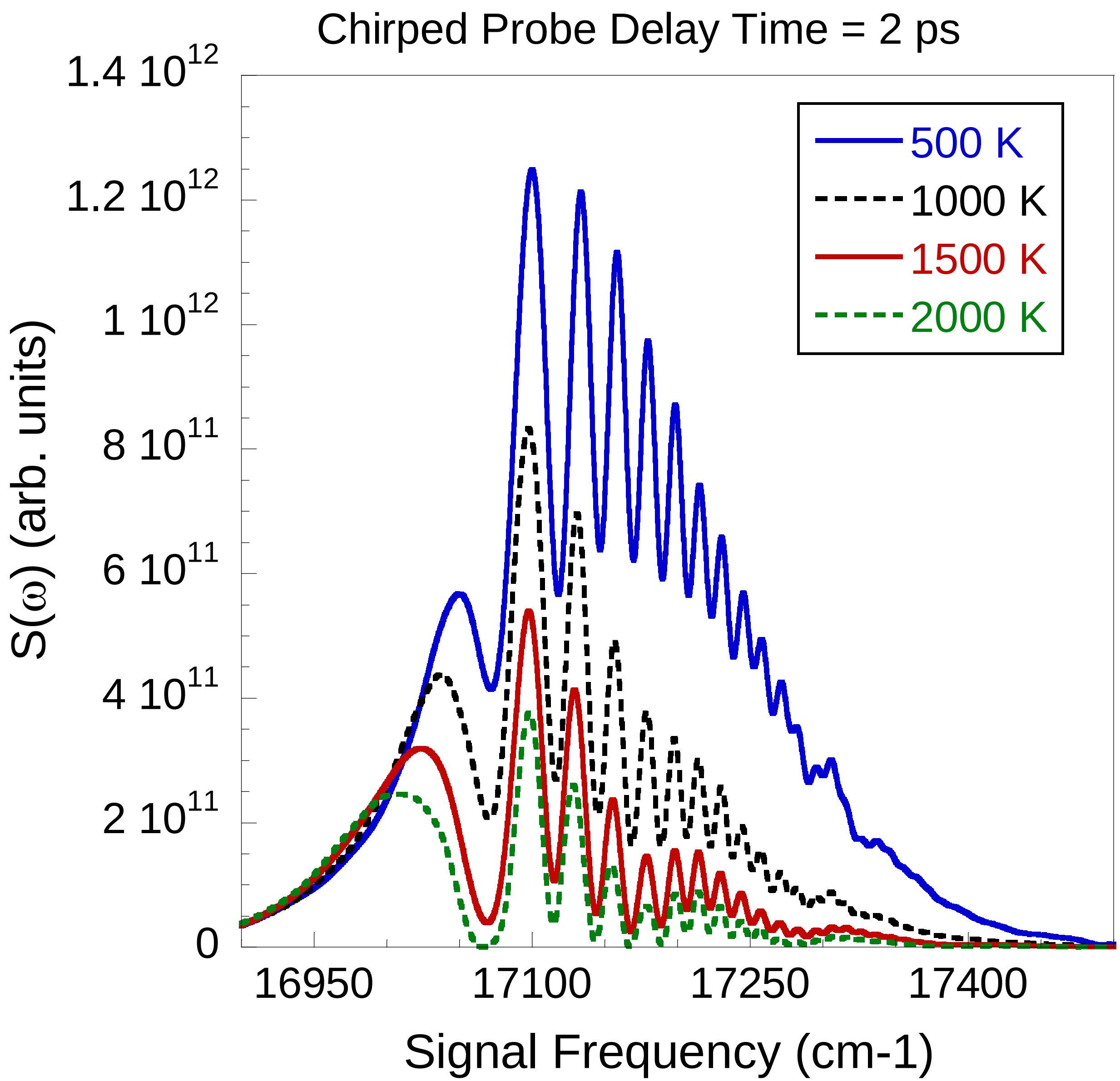
Temperature Measurements in Flames



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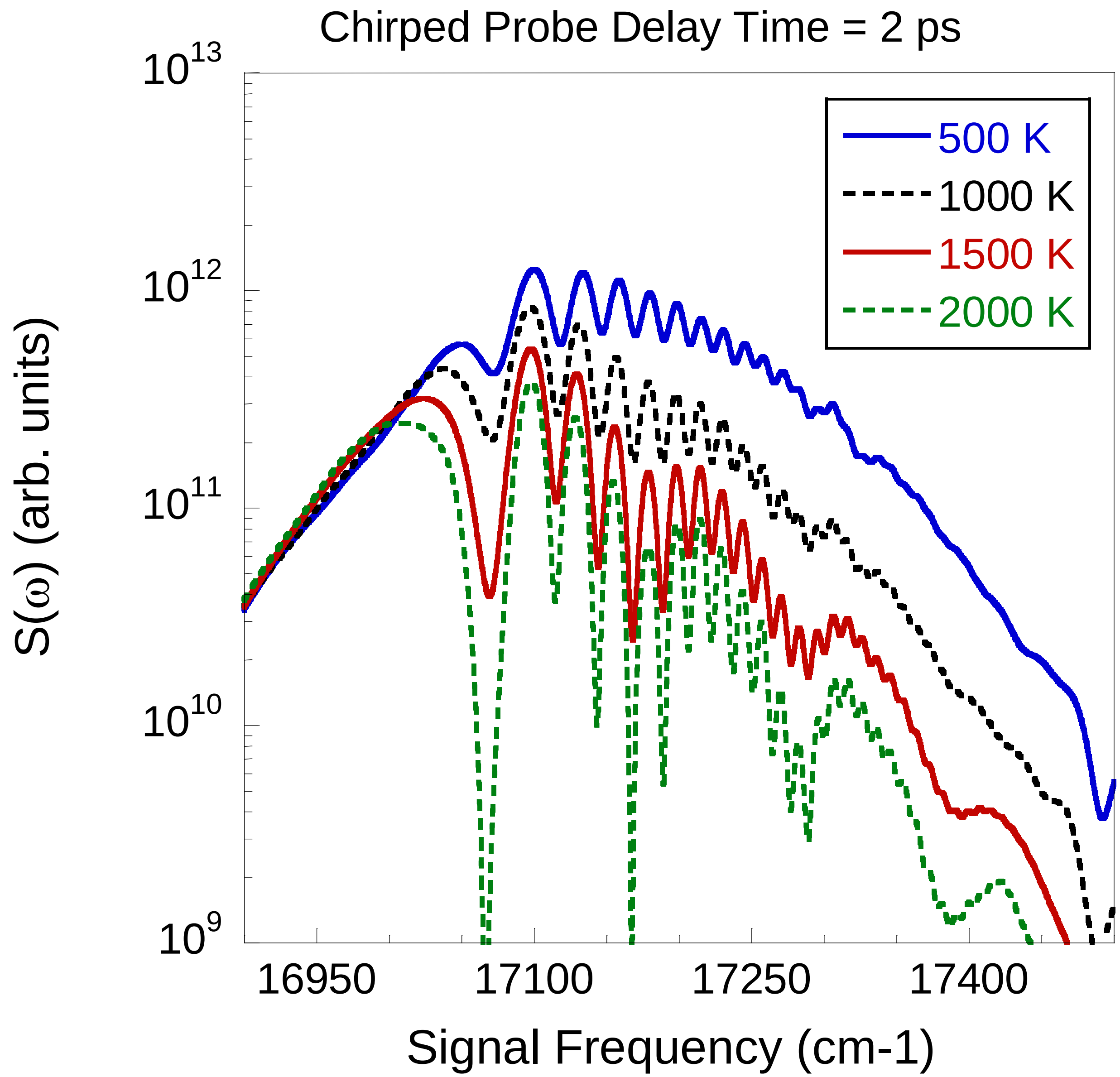
Temperature Measurements in Flames



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Numerical Model of fs CARS in N_2

- **A model of the CARS process in nitrogen based on direct numerical integration of the time-dependent density matrix equations has been developed [Lucht et al., Journal of Chemical Physics, 127, 044316 (2007)].**

- **Model is nonperturbative and is based on direct numerical integration of the time-dependent density matrix equations.**

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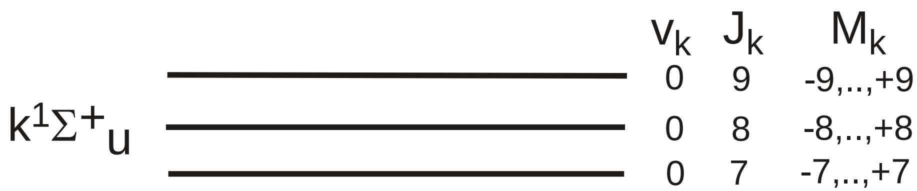
Single-Laser-Shot Fs CARS Measurements

- Most significant difference for fs CARS compared to ns CARS is the potential for data rates of 1-100 kHz as compared to 10-30 Hz for ns CARS.

Numerical Model of N_2 CARS



$k^1\Sigma^+_u$-$X^1\Sigma^+_g$ transition strength varied to match N_2 Raman cross section