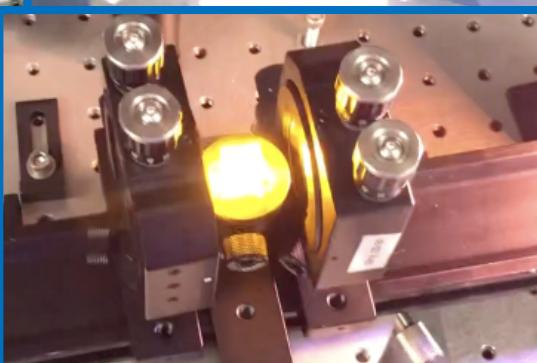
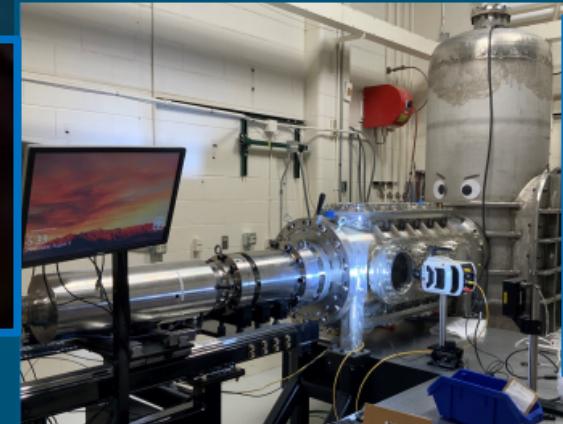
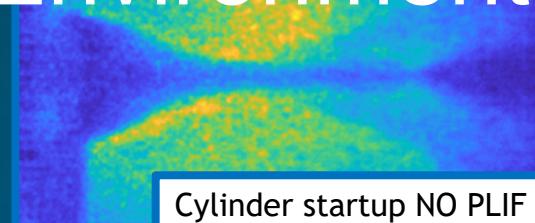


# CARS Thermometry and Species Measurements in Extreme-Temperature Environments



Sandia  
National  
Laboratories

Sean P. Kearney  
*Engineering Sciences Center*  
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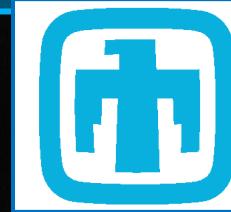


Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

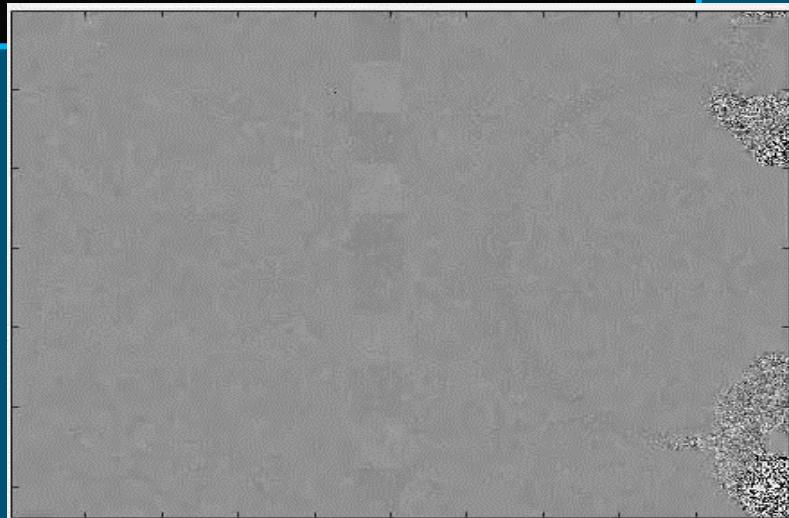
## 2 Our Motivation: High Temperature Materials Test Environments



### Free-Piston Reflected Shock Tunnel

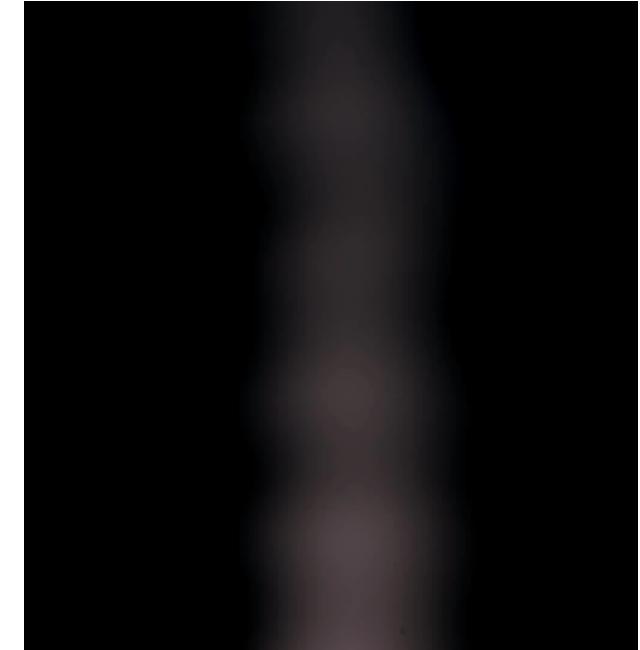
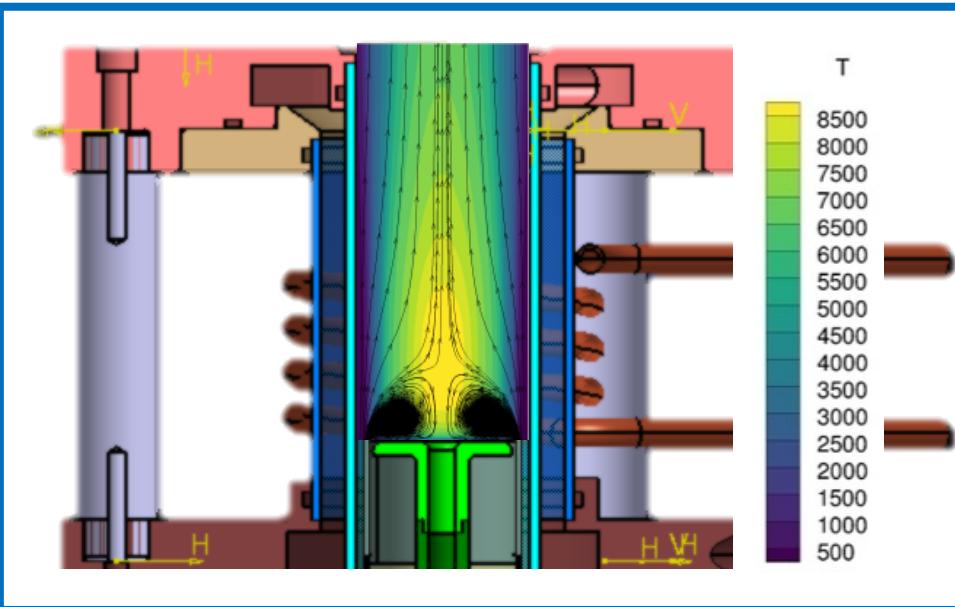
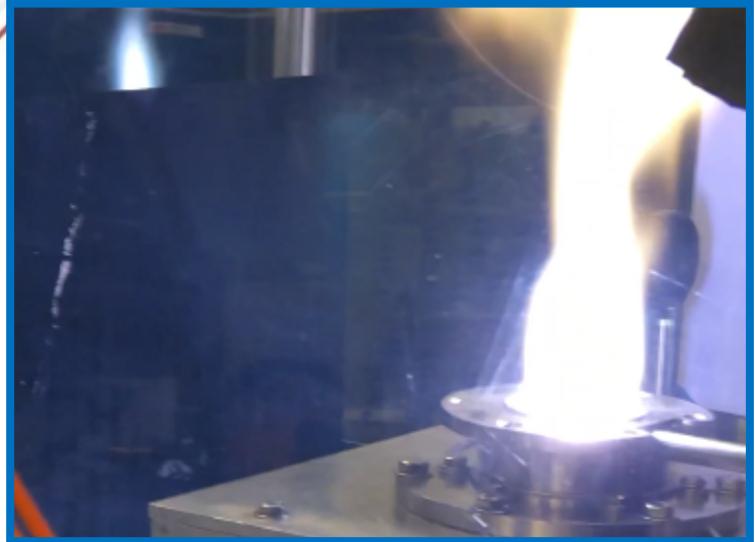


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National  
Laboratories



### Inductively Coupled Plasma Torch

# N<sub>2</sub> CARS Thermometry in IC Plasma Torch at UT-Austin



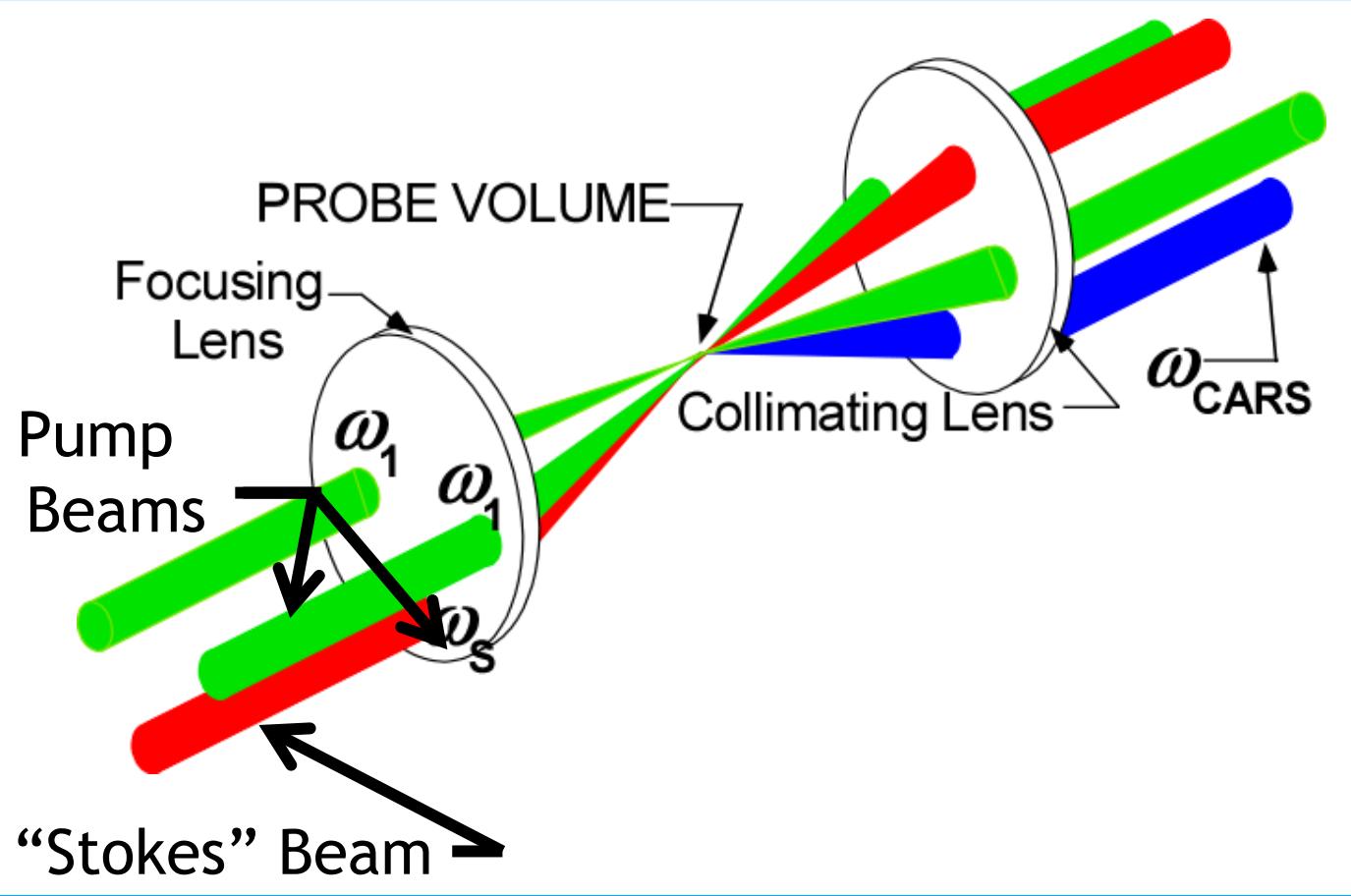
## Inductively Coupled Plasma Torch

- Source for testing of hypersonic TPS materials
- “Chemically clean” – electrodes isolated from flow
- Realistic temperatures (T > 6000 K)
- Low flow rate – cannot match flight conditions

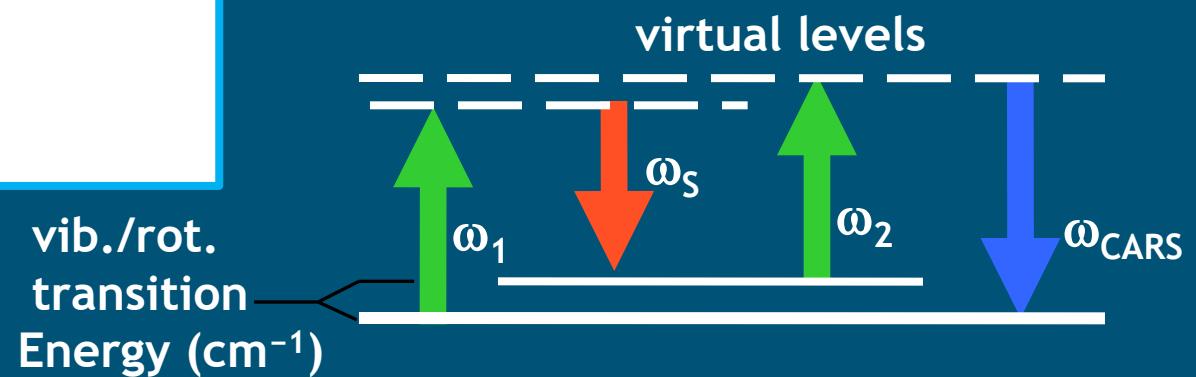
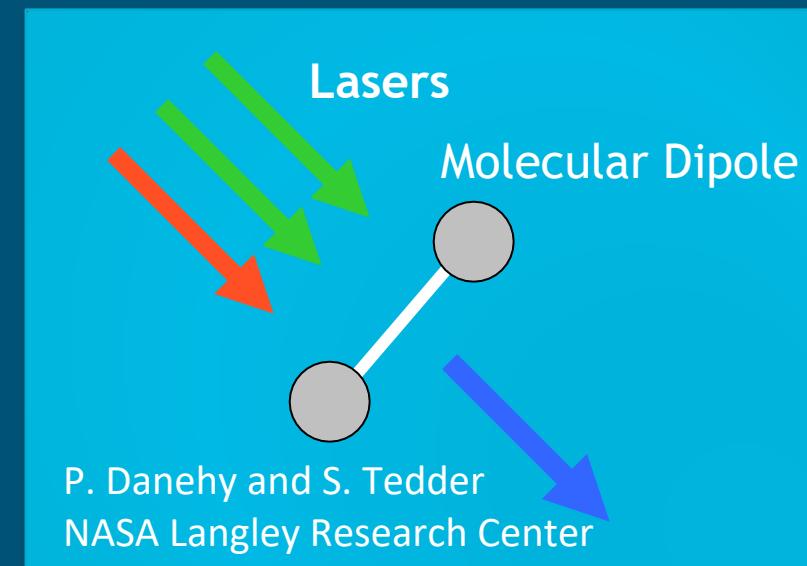
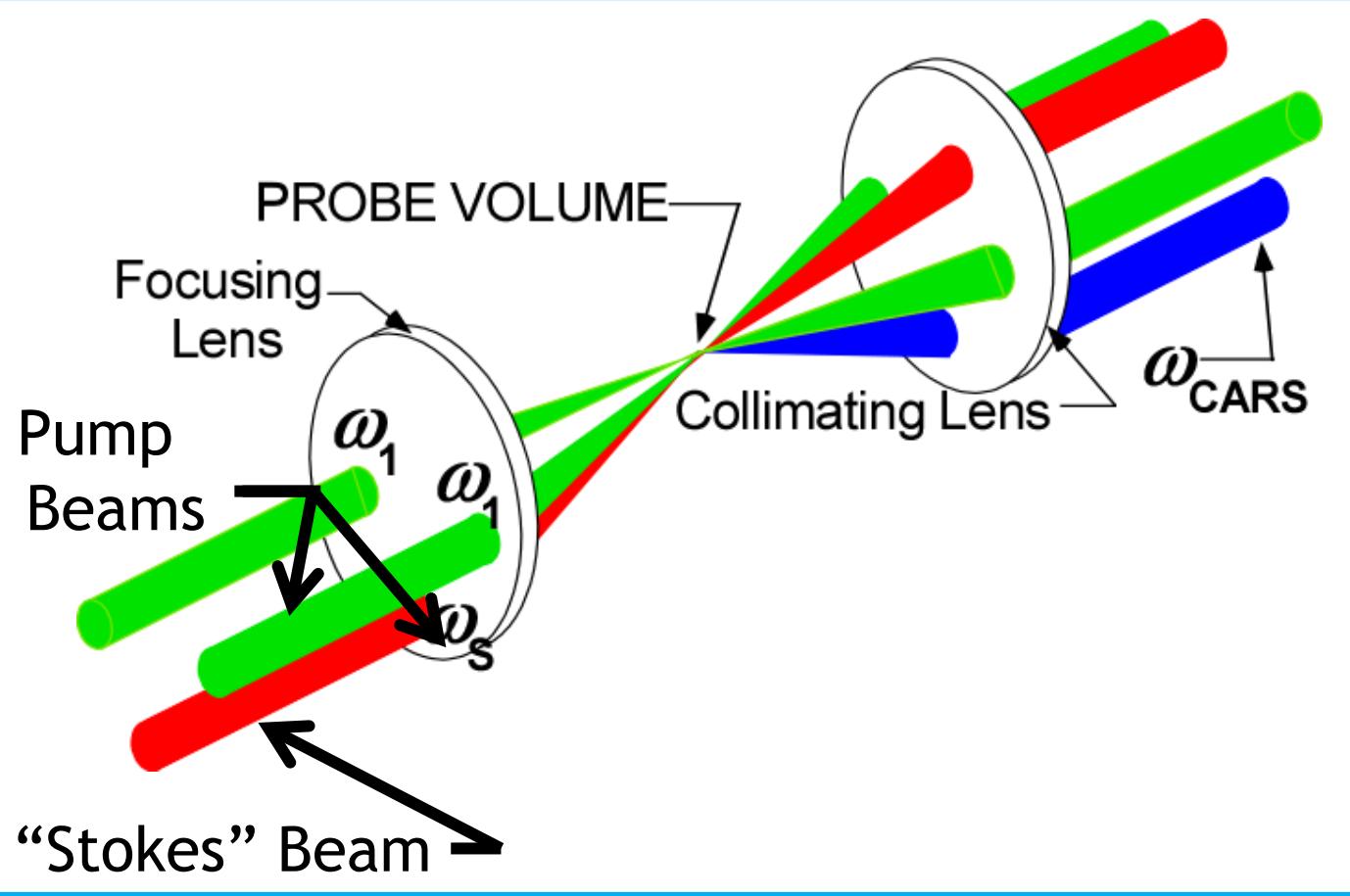
Collaboration with DOE/PSAAP Center at UT



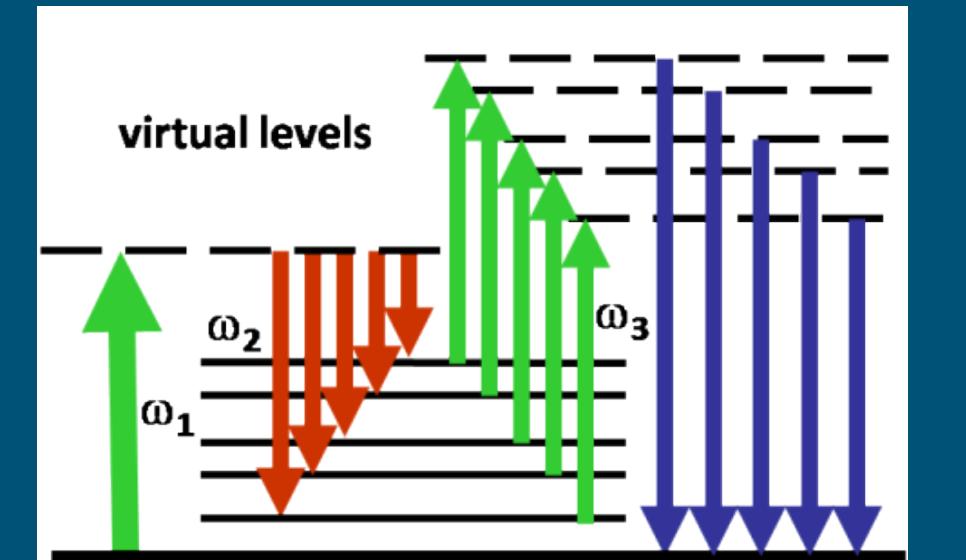
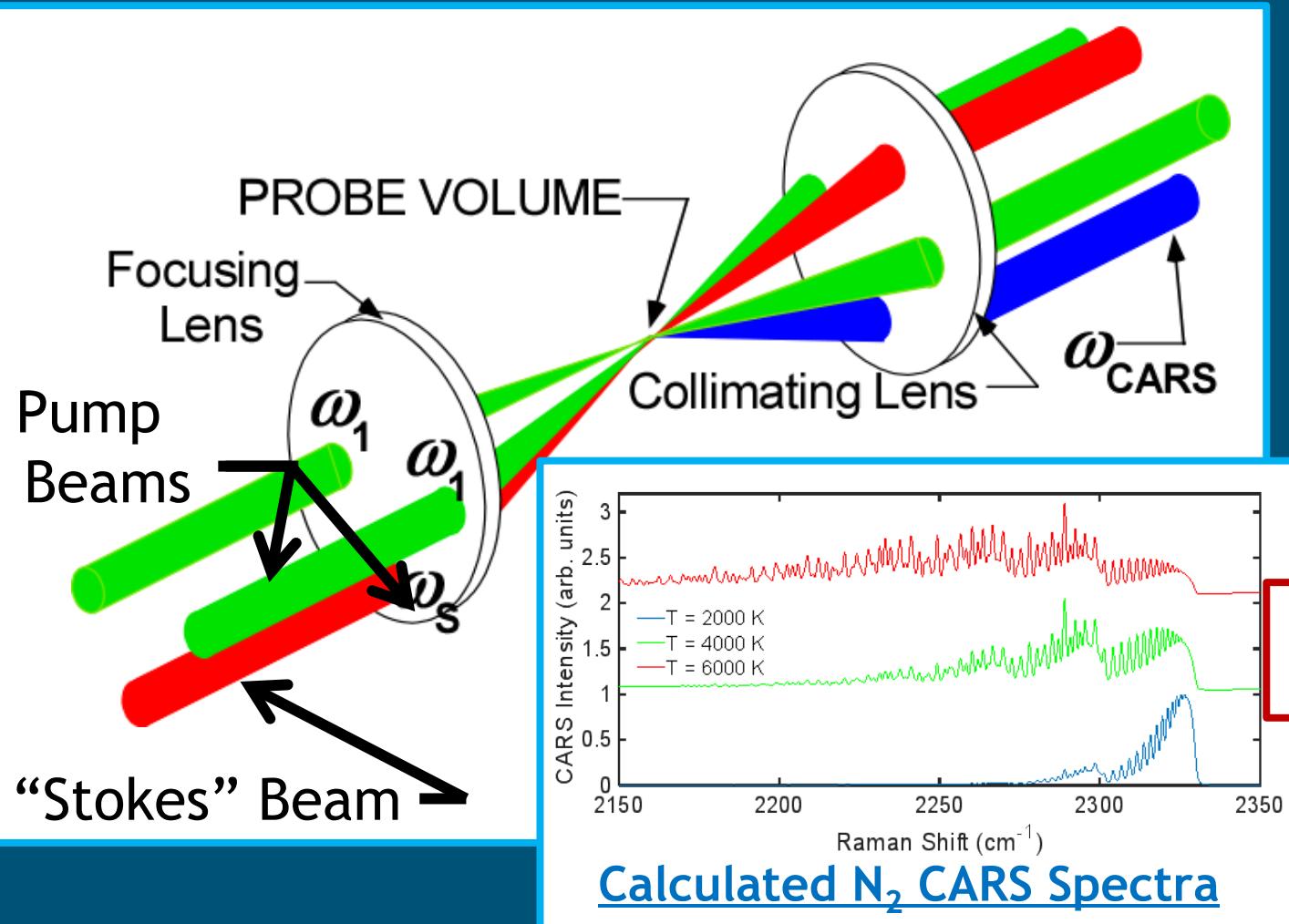
# Coherent anti-Stokes Raman scattering (CARS)



# Coherent anti-Stokes Raman scattering (CARS)



# Coherent anti-Stokes Raman scattering (CARS)



Broadband Stokes Source Pumps Multiple Raman transitions simultaneously

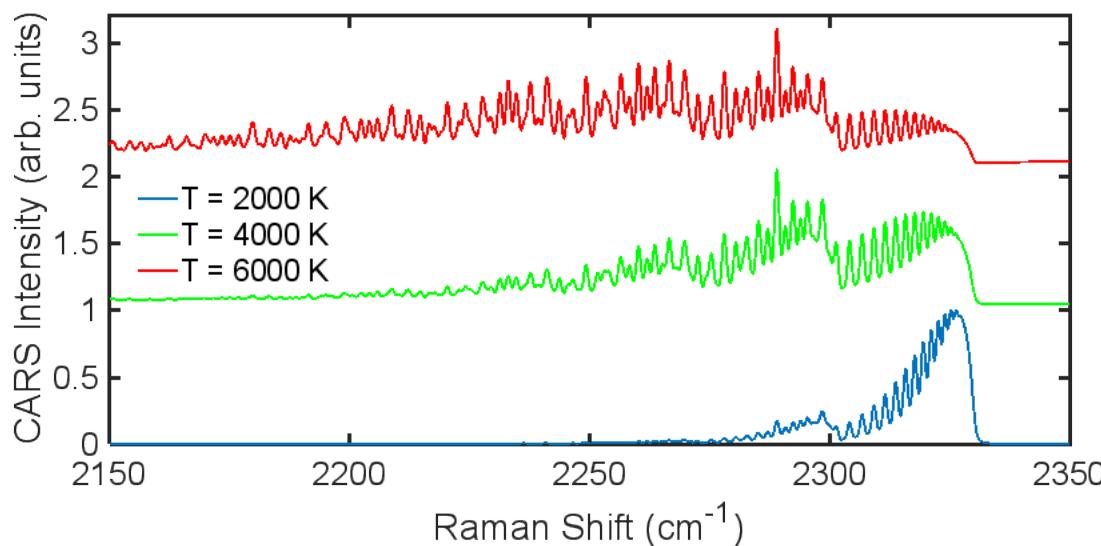
# IC Plasma Torch N<sub>2</sub> Thermometry

## "Conventional" CARS Instrument

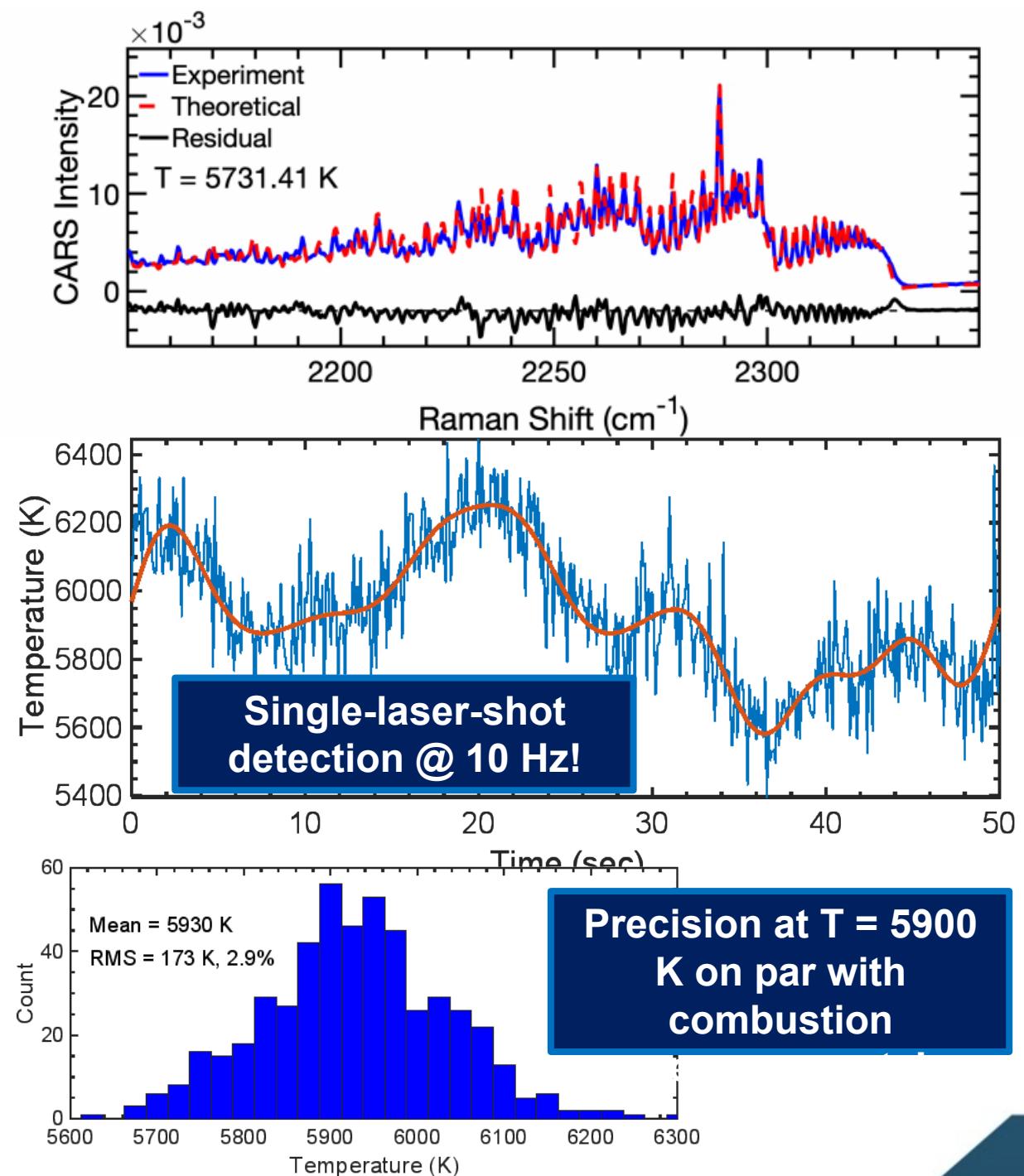
- ~10-ns laser pulses at 10-Hz
- Broadband dye laser for Stokes source

## Key technical challenges

- Very high background luminosity
- Extreme temperatures, some of the highest ever measured with this technique

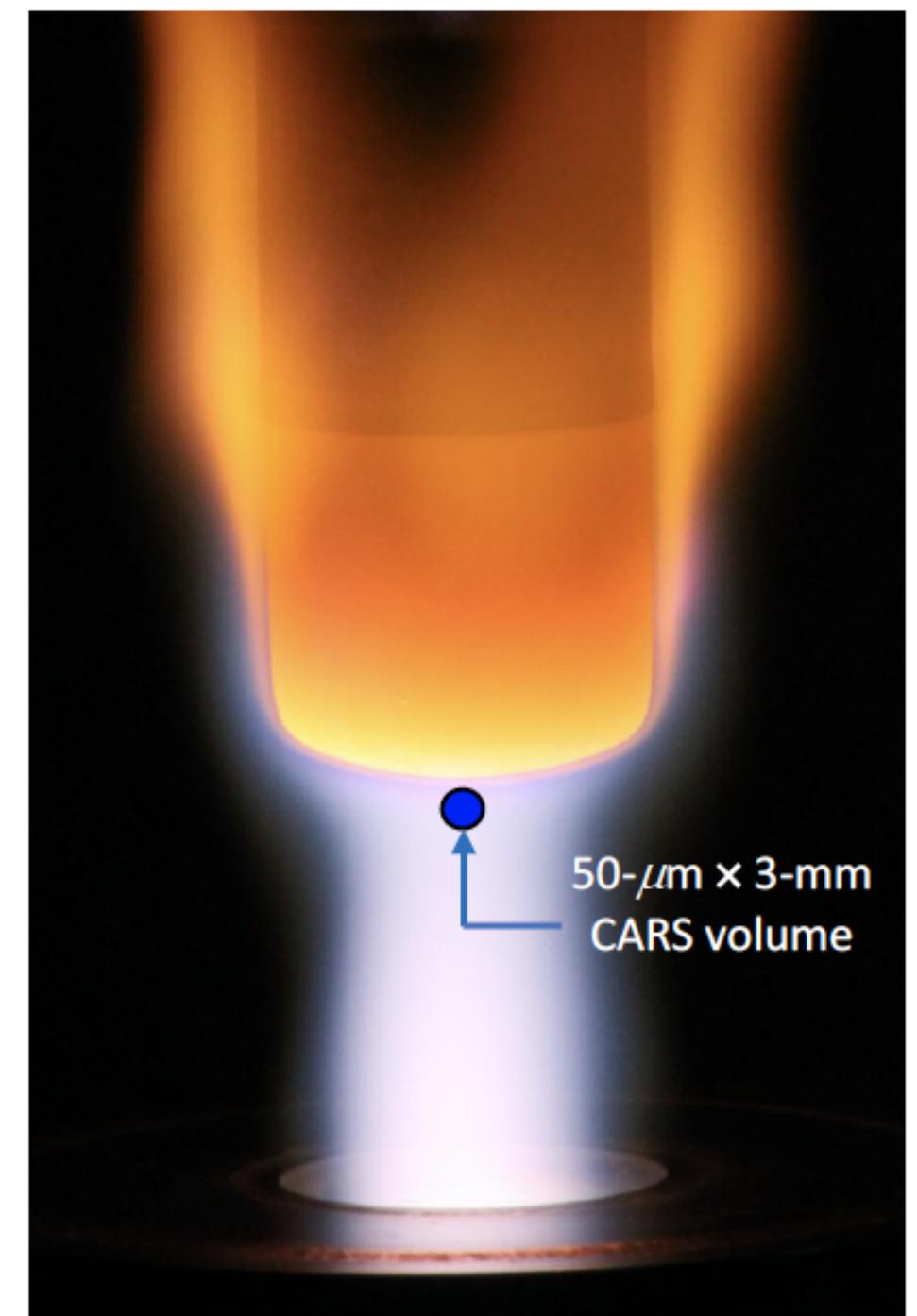
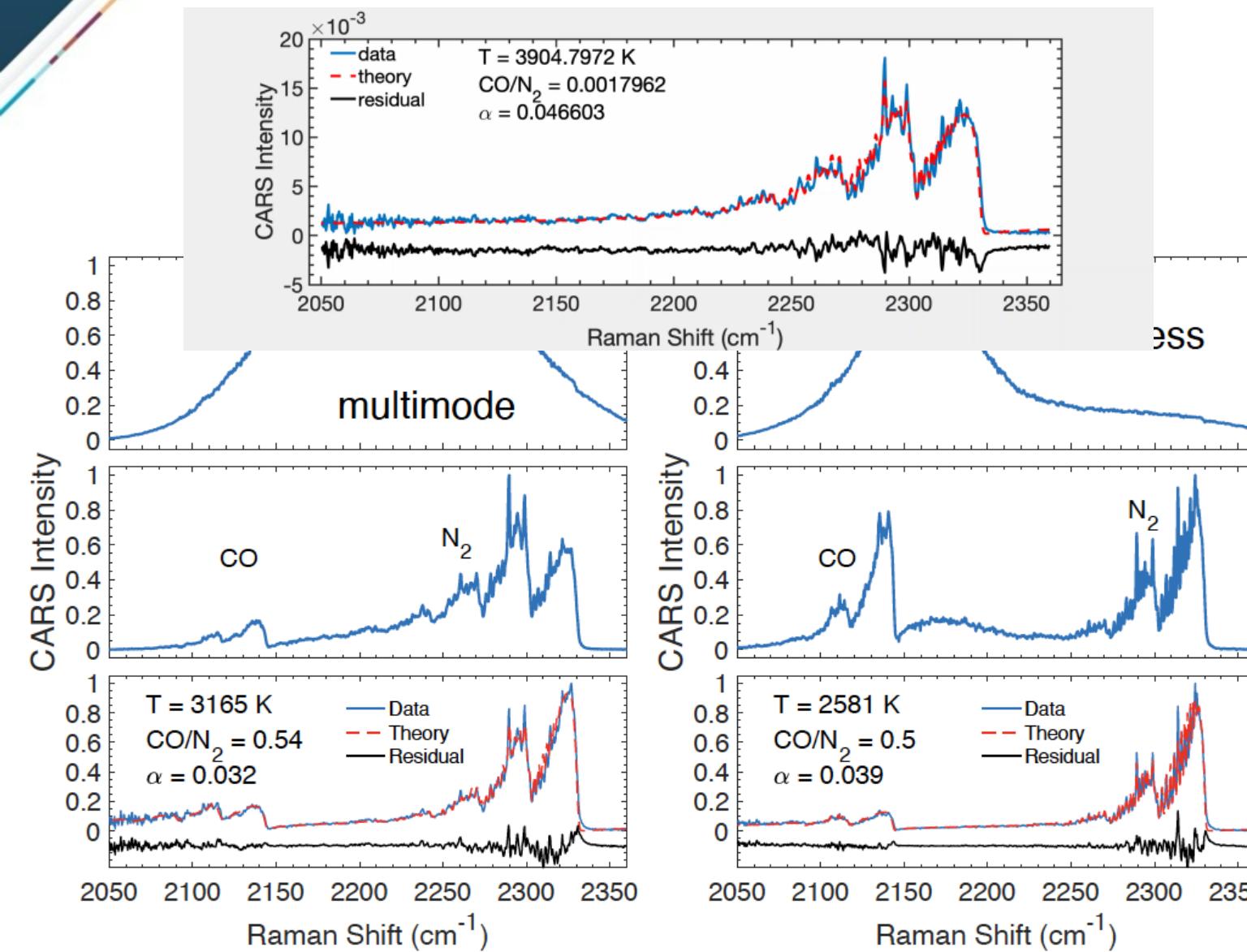


## Calculated N<sub>2</sub> CARS Spectra



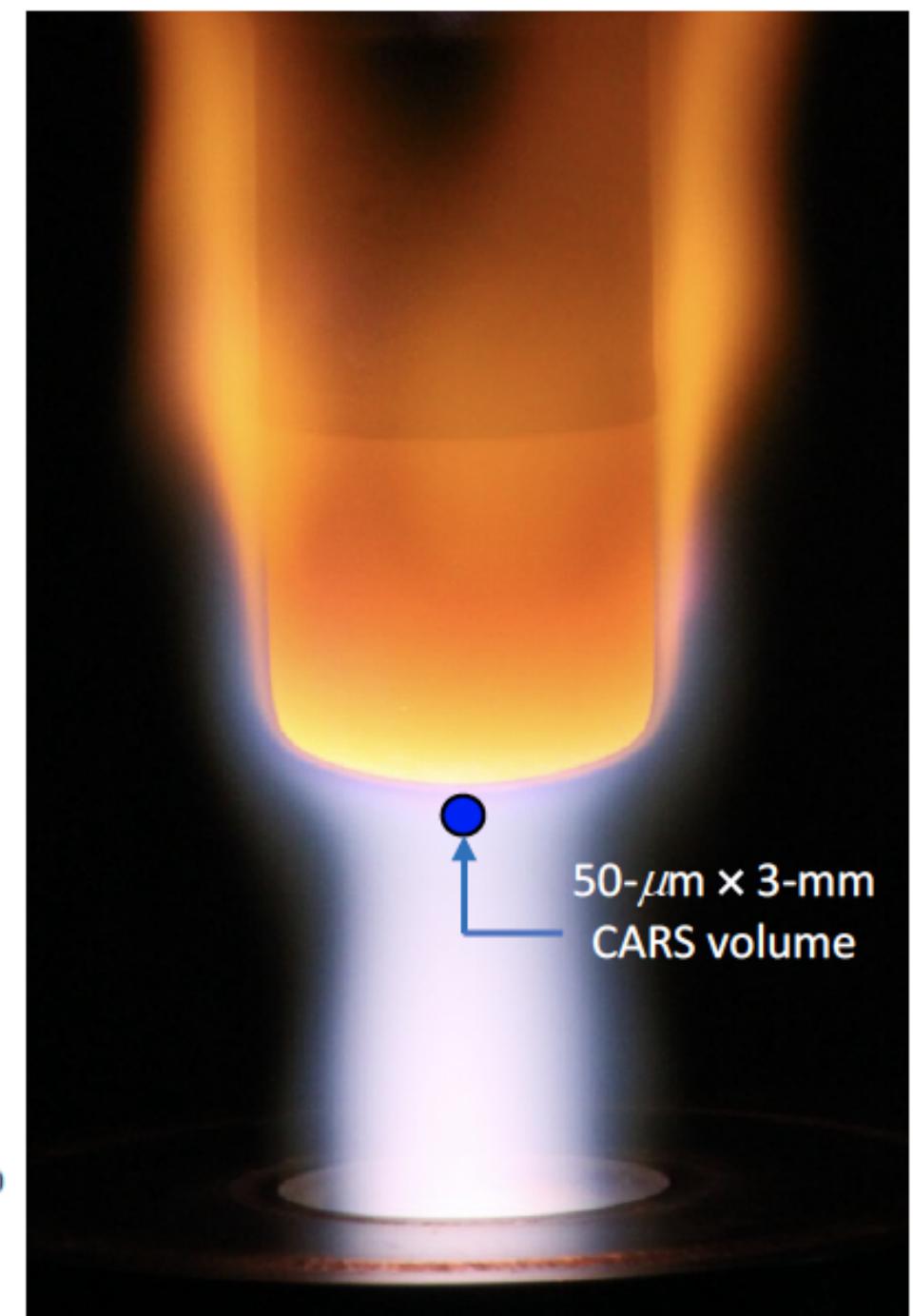
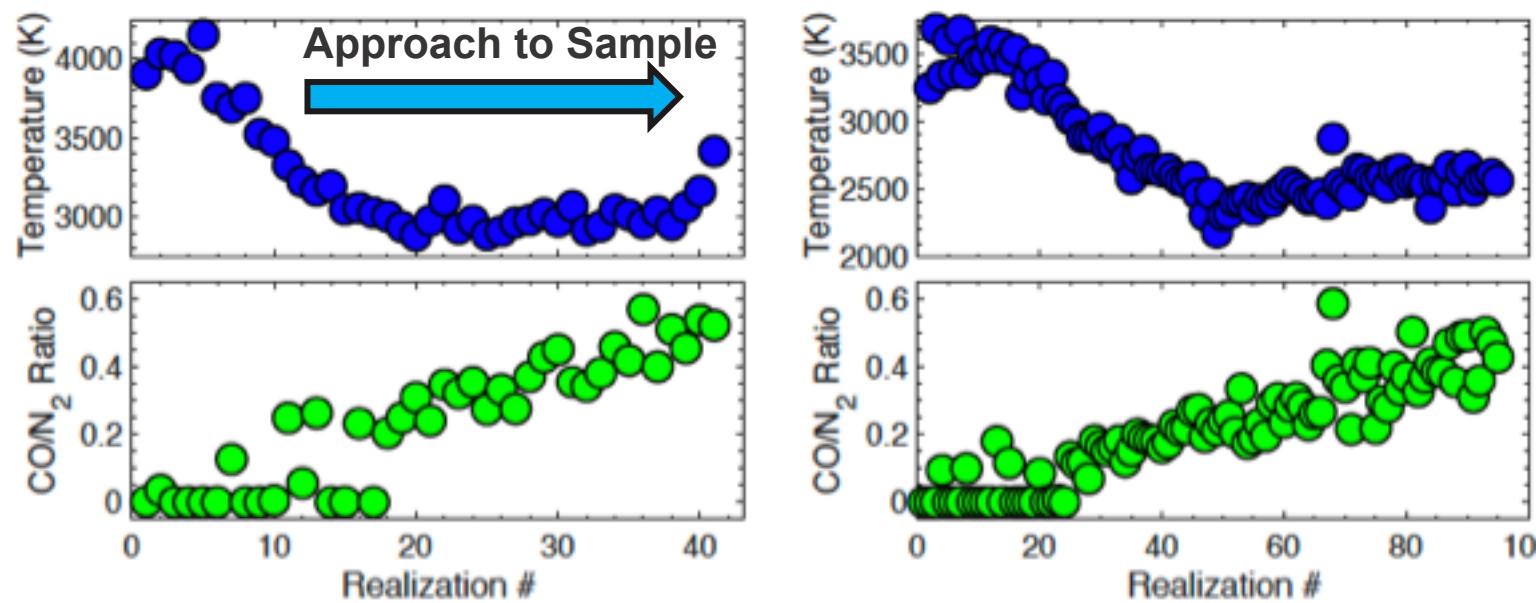
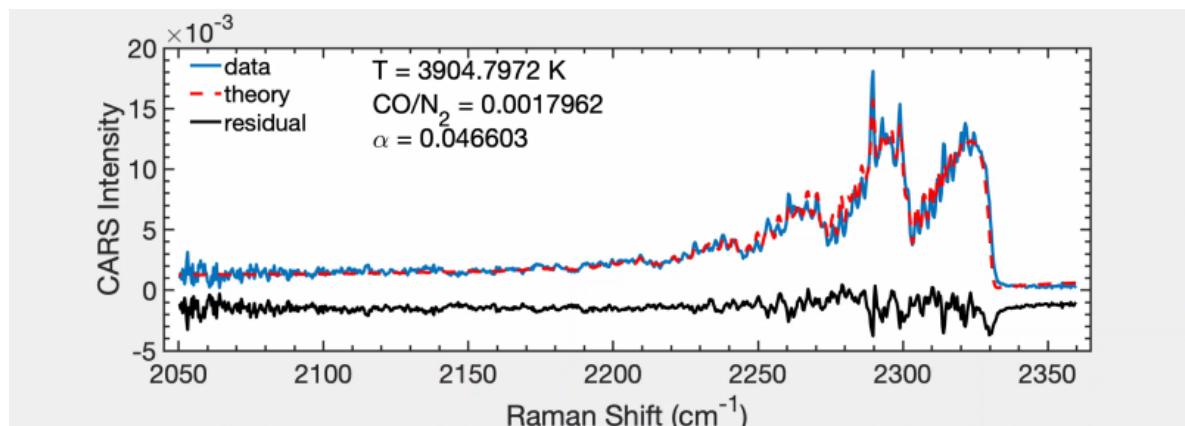


# CO/Temperature Measurements in High-T Reaction Layer

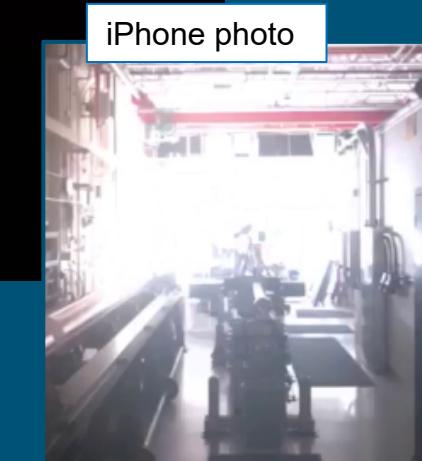
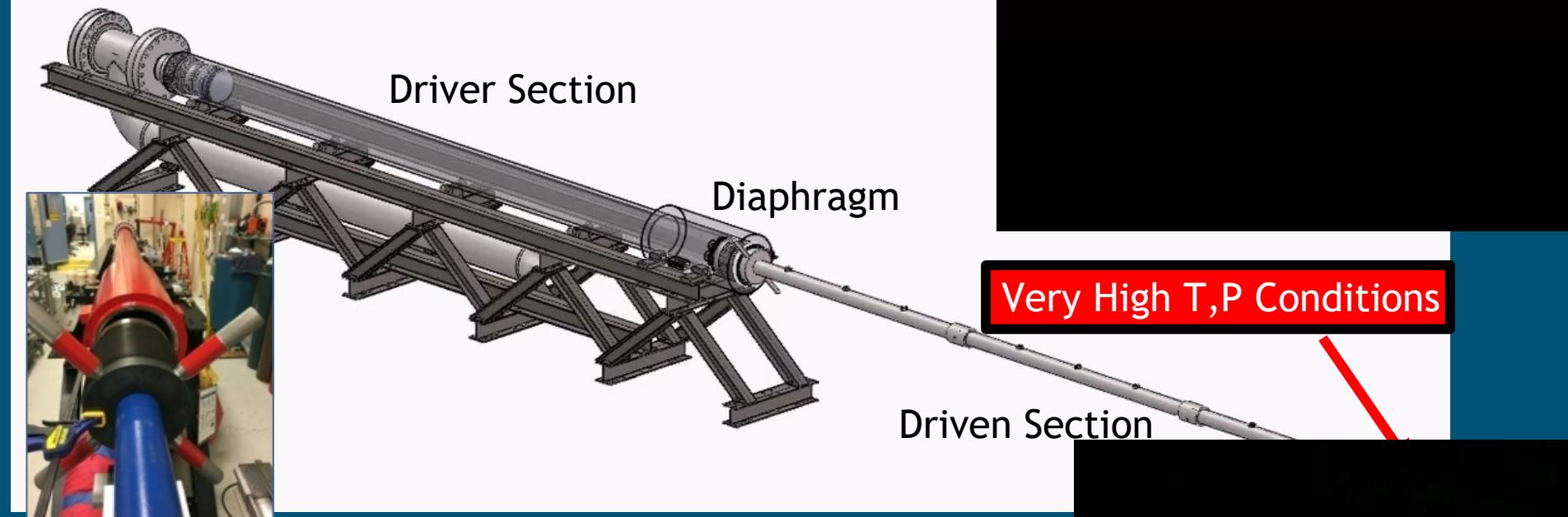




# CO/Temperature Measurements in High-T Reaction Layer



# Sandia Free-Piston-Driven High-Temperature Shock Tube (HST)



Temperature (K)

$$T_1 = 292 \text{ K}, P_1 = 0.1 \text{ bar}$$

$$T_2 = 2090 \text{ K}, P_2 = 1.1 \text{ bar}$$

$$T_5 = 4013 \text{ K}, P_5 = 10 \text{ bar}$$

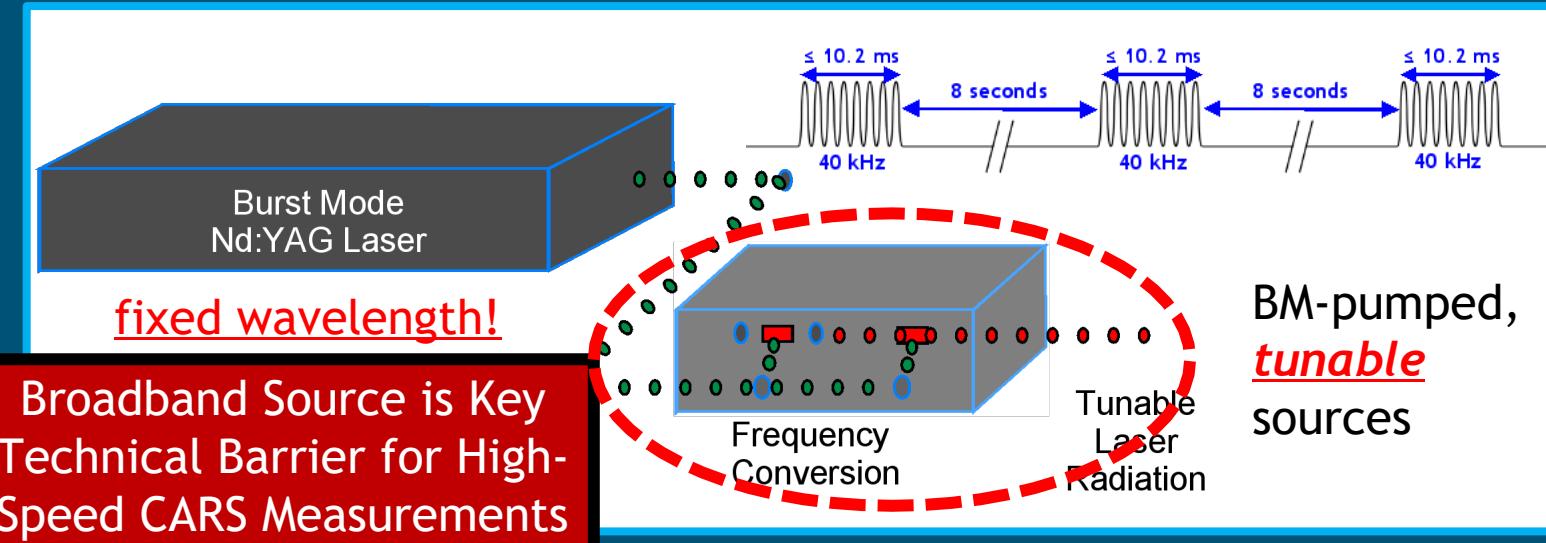
Contact Surface

**Test time = 1 ms or less!**

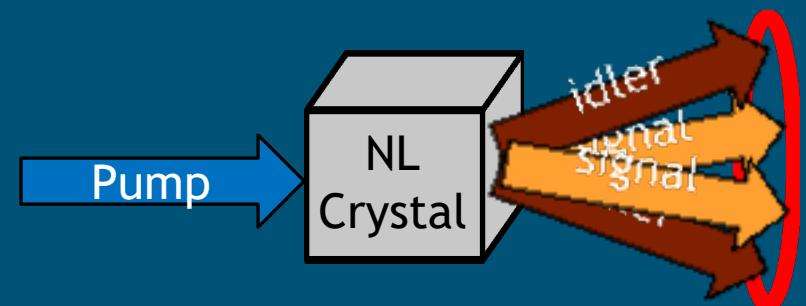
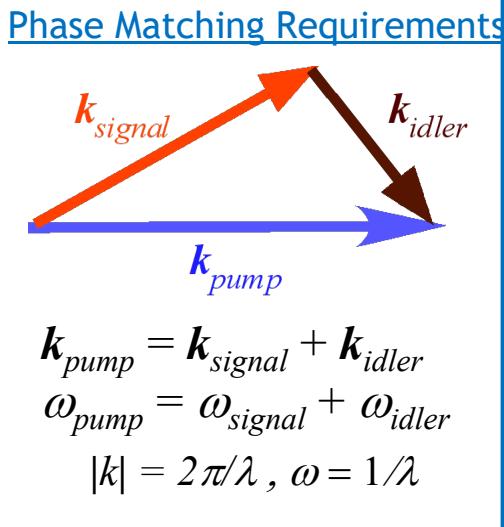
# Picosecond Pulse-Burst CARS at 100 kHz for HST Application



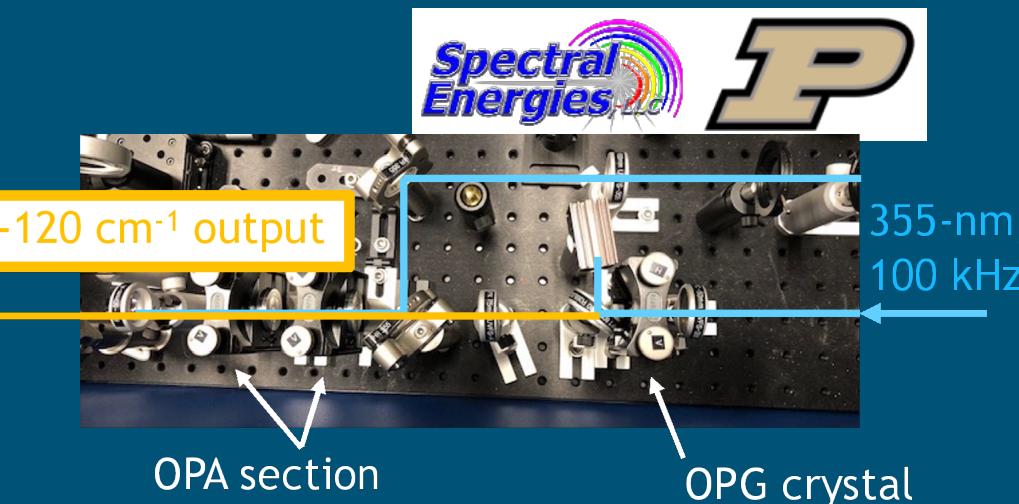
- Burst-mode lasers have allowed experimentalists to access high-speeds (10s to 100s of kHz)
- While powerful, these systems are not wavelength tunable—this prohibits application of ***chemically specific*** imaging and spectroscopic tools



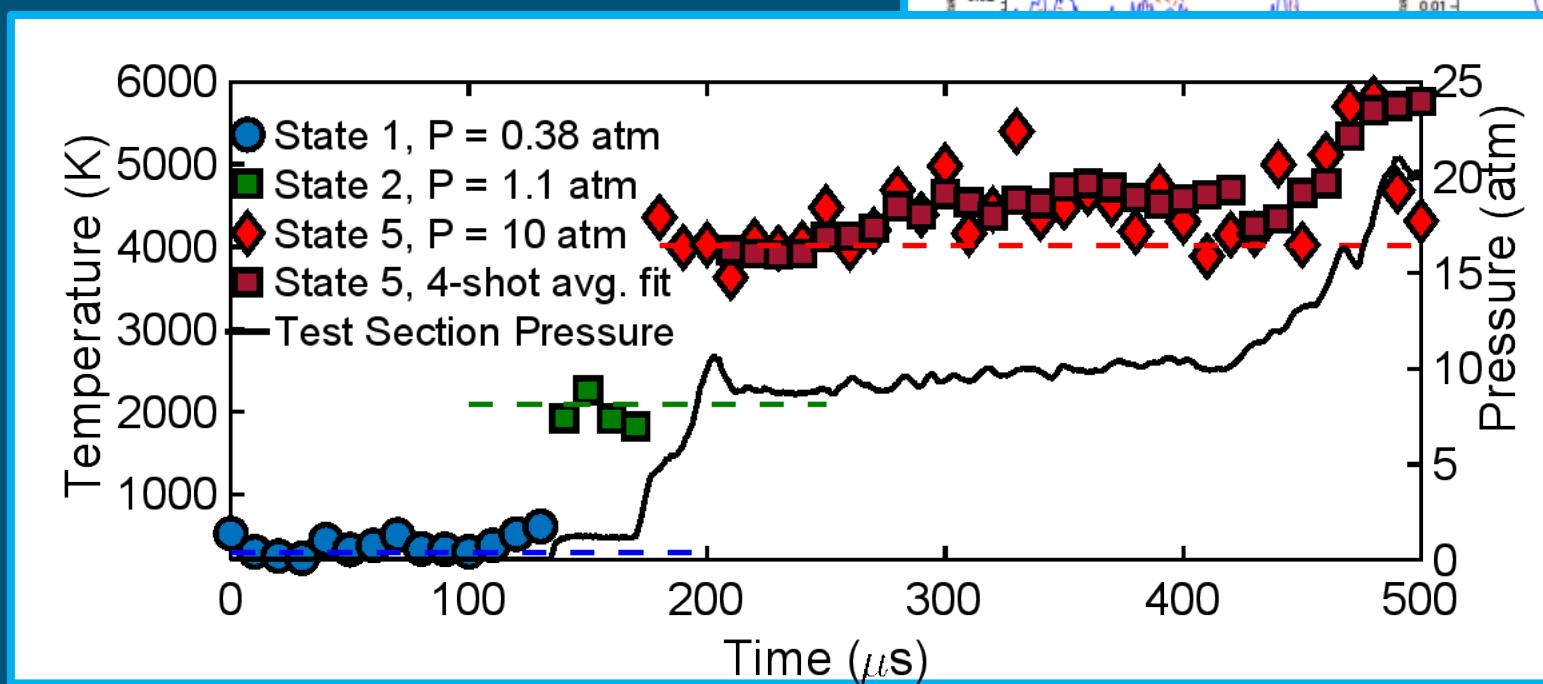
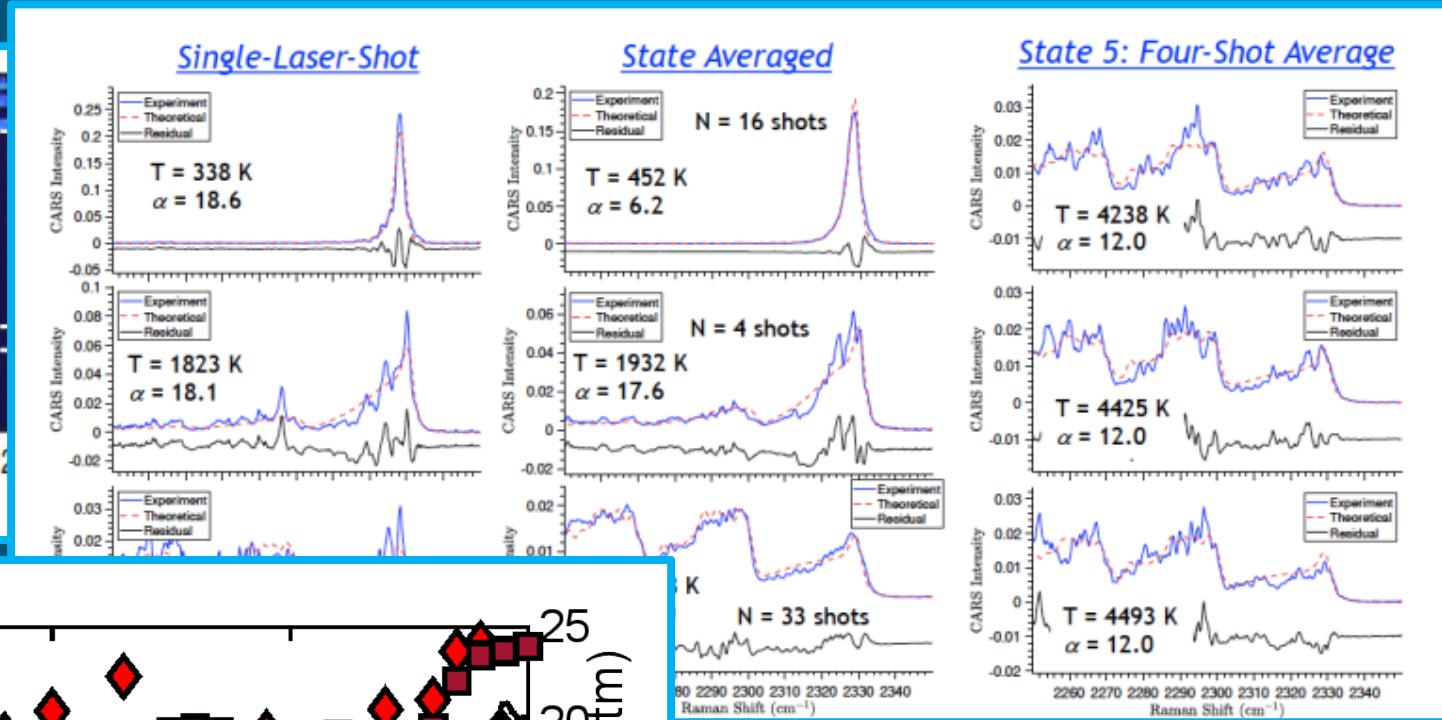
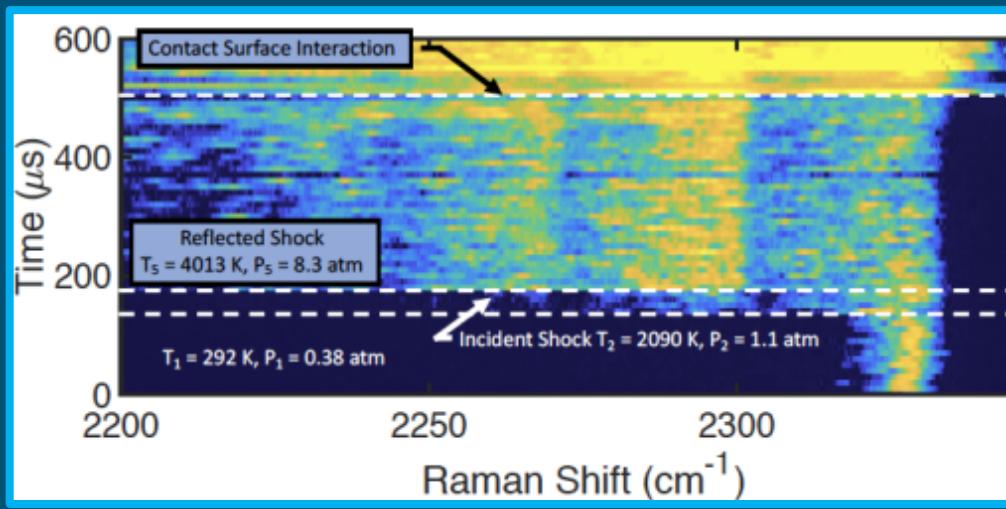
## Broadband Picosecond Optical Parametric Generator/Amplifier (OPG/OPA)



Picosecond OPG = Enabling Technology!



# 100-kHz Pulse-Burst CARS in the Sandia Free-Piston Shock Tube

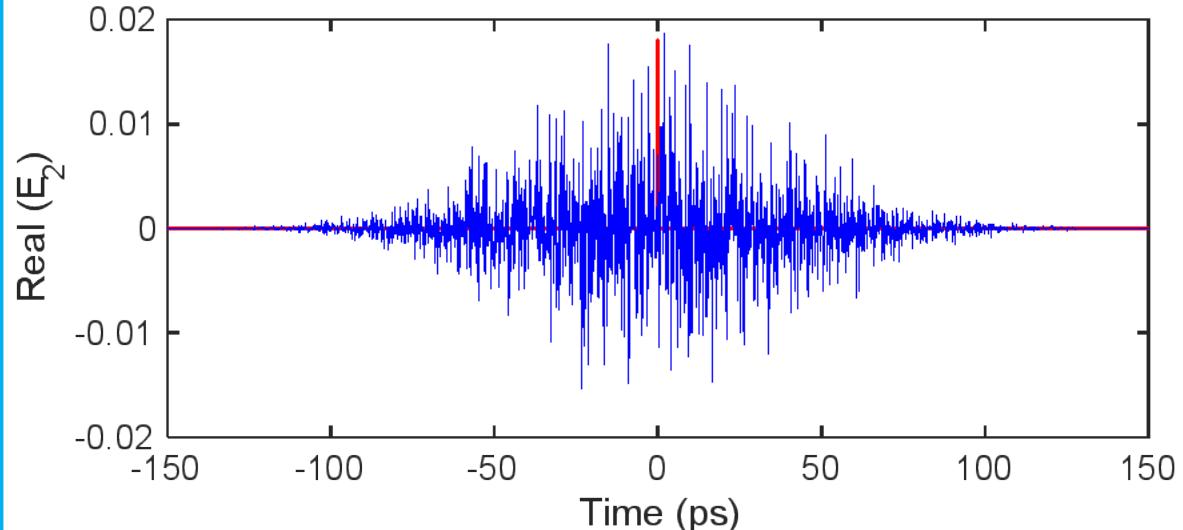


# Picosecond CARS comes with a single-shot noise penalty

Dominant source of CARS noise is the quality of the broadband pulse

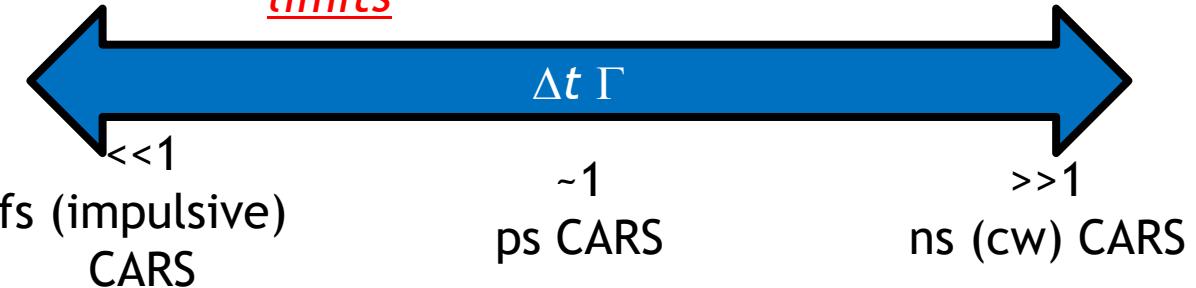
Time-Bandwidth Product - Fourier-Transform Limit

$$\Delta t [\text{ps}] \Delta \omega [\text{cm}^{-1}] \geq 14.67$$



- To achieve sufficient bandwidth for CARS, pulse must exhibit ~150-fs features - inherently noisy!

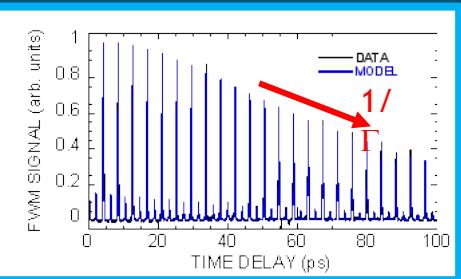
Picosecond CARS lies between cw and impulsive limits



Transform limited (low noise)

Time averaged

- $\Delta t \sim 50-100 \text{ ps} \sim 1/\Gamma \rightarrow$  very little averaging in the Raman process



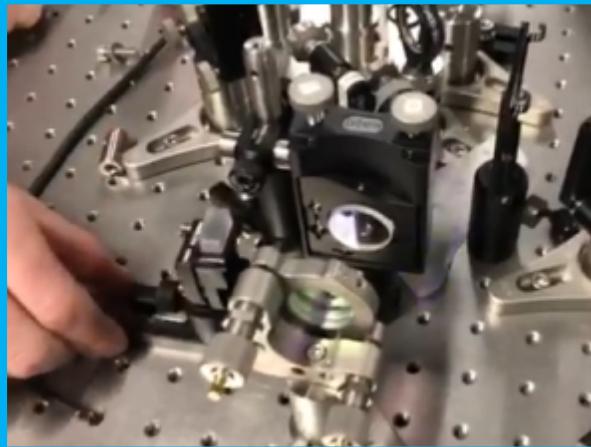
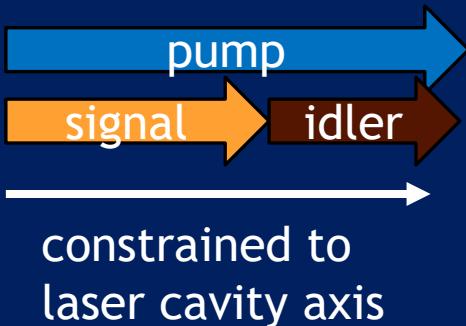
# Nanosecond Burst-Mode CARS via *Noncolinear* Optical Parametric Oscillator

14



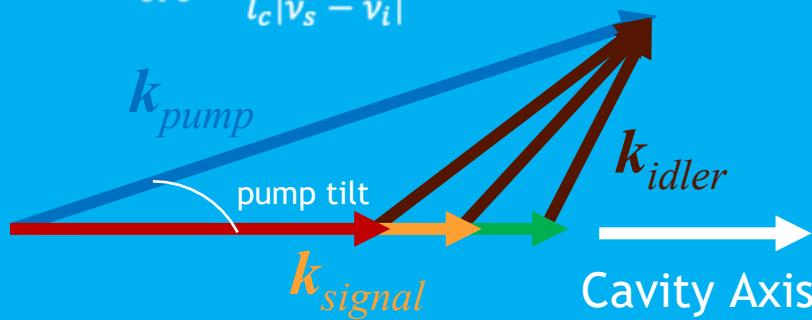
Conversion of low-intensity nanosecond pulses requires laser-cavity gain

Most OPOs are co-linear to satisfy phase-matching constraint



OPO Bandwidth is enhanced by matching group velocities of signal and idler waves

$$FWHM_{OPO} = \frac{c}{l_c |v_s - v_i|}$$



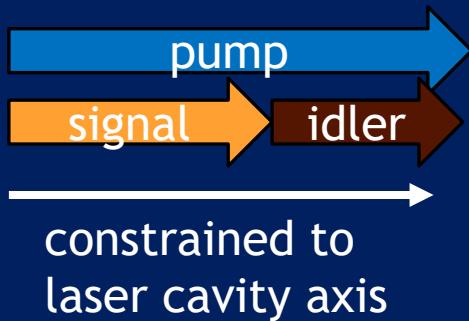
# Nanosecond Burst-Mode CARS via *Noncolinear* Optical Parametric Oscillator

15



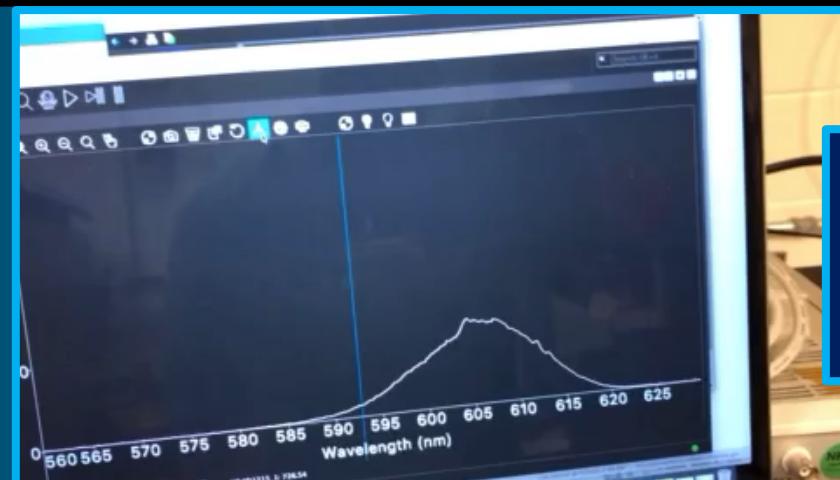
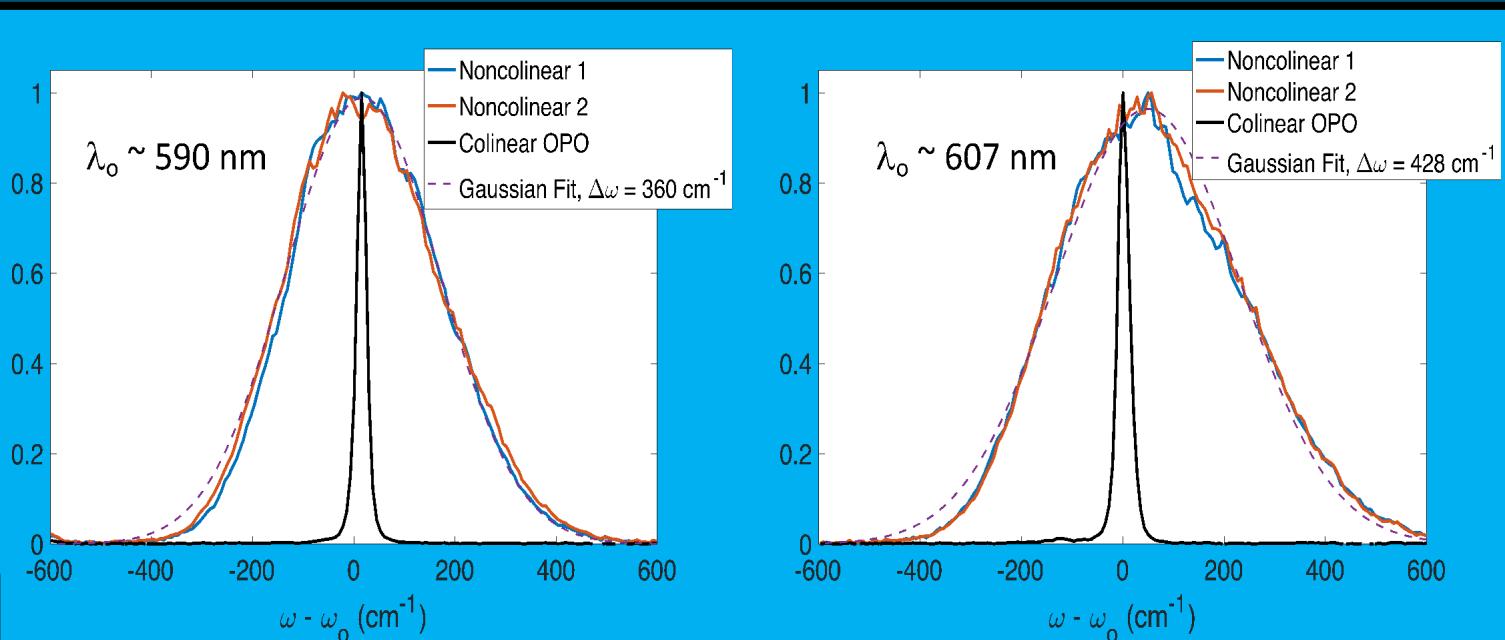
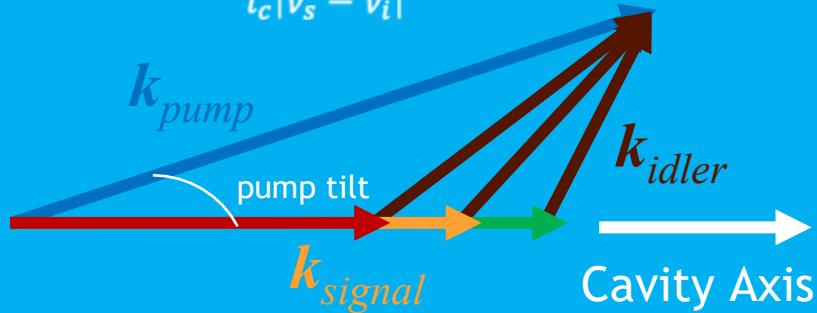
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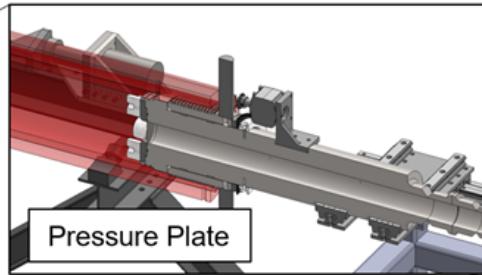


iPhone movie of NOPO spectrum tuning with 10-Hz pump laser

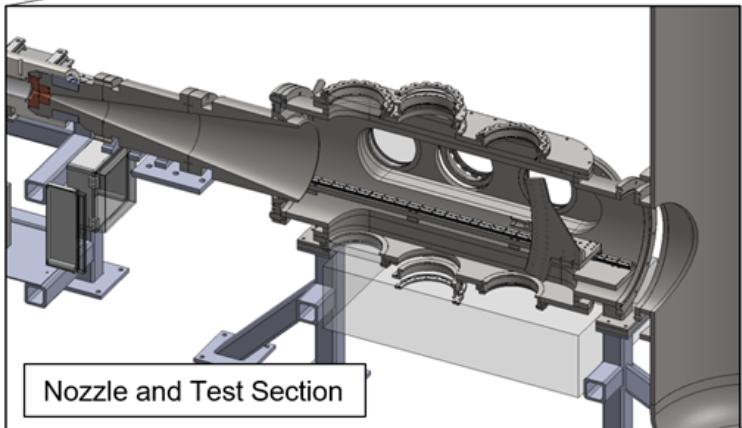
# Hypersonic Shock Tunnel (HST) Provides the Reentry Flight Environment



Lynch et al. 2022



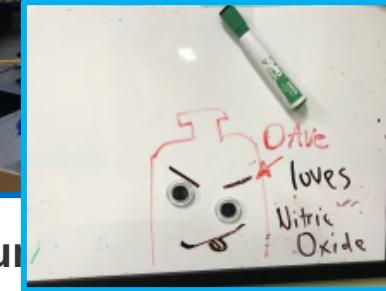
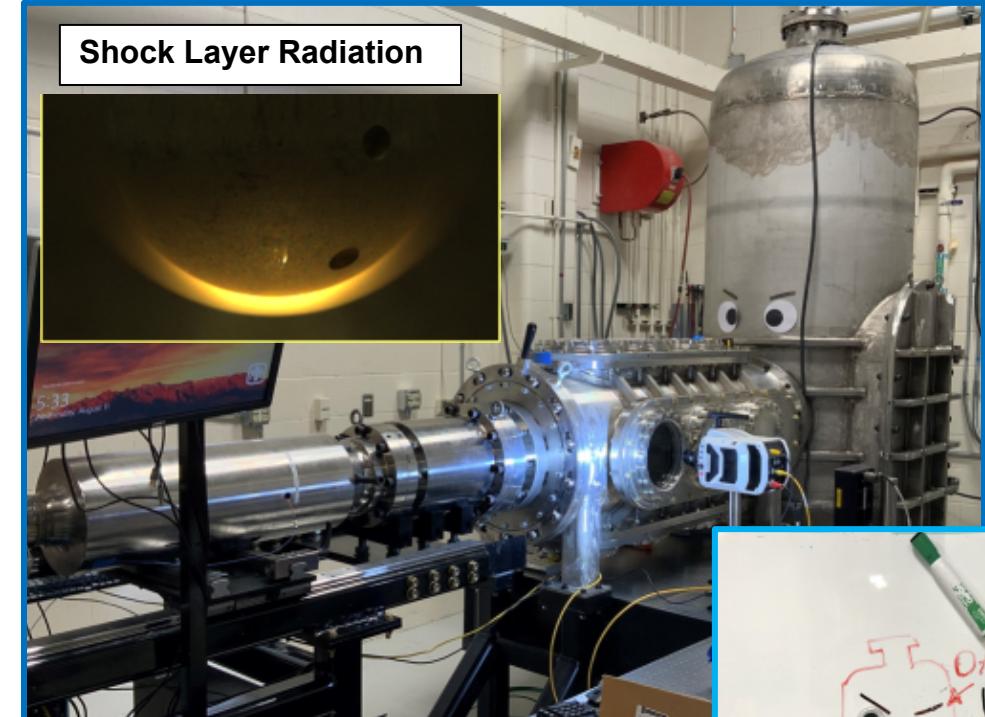
Pressure Plate



Nozzle and Test Section

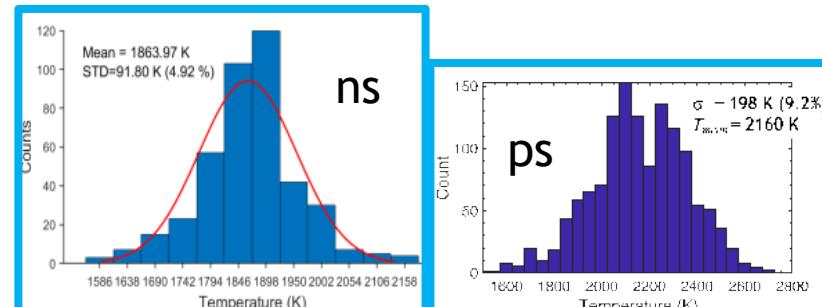
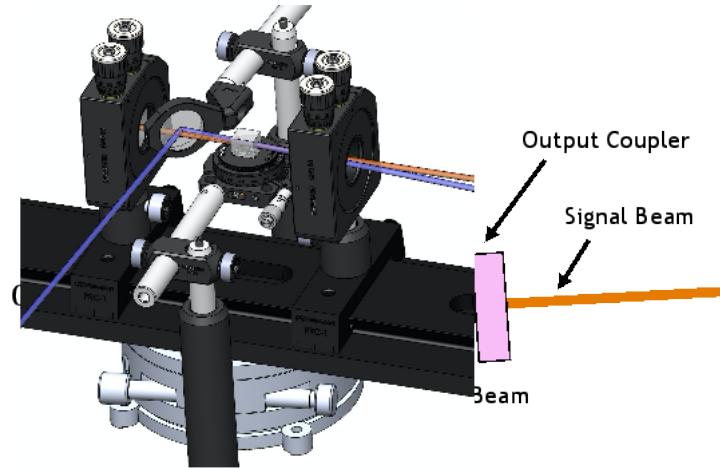
Vacuum Tank

- Multimode: can run as **shock tube** or **shock tunnel**
- First experiments at **Mach 8** flight enthalpy
  - Pressure altitude of 42 km (136 kft).
  - Stagnation temperature  $\approx 3700$  K
- **Research applications:**
  - Aerothermodynamics including nonequilibrium
  - Thermal protection system (TPS) materials



First shot of tunnel in August 2021!

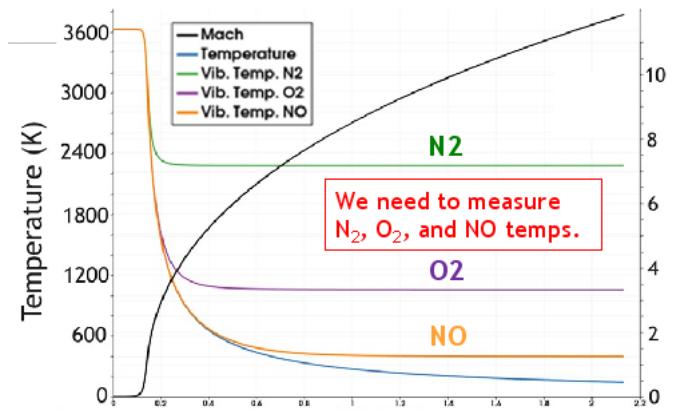
# Improved high-speed thermometry: 100-kHz nanosecond CARS



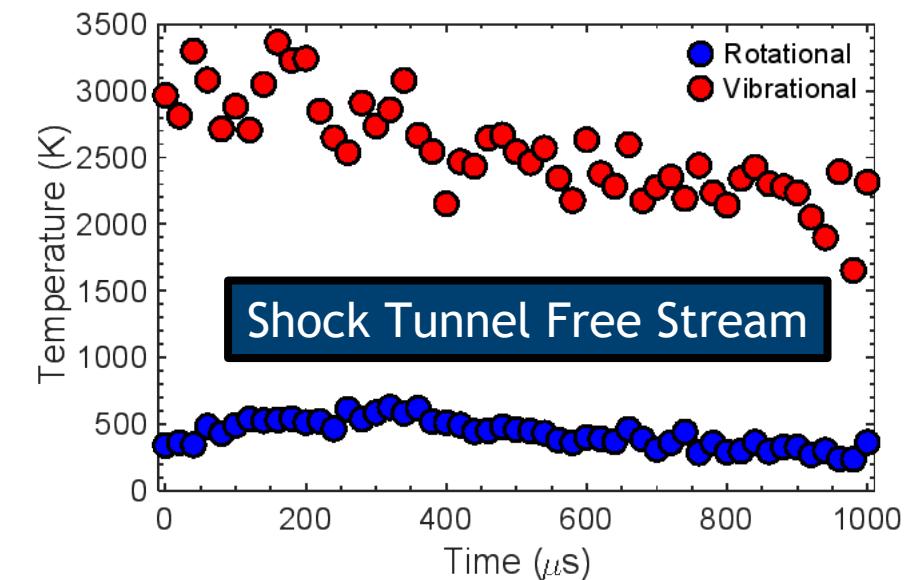
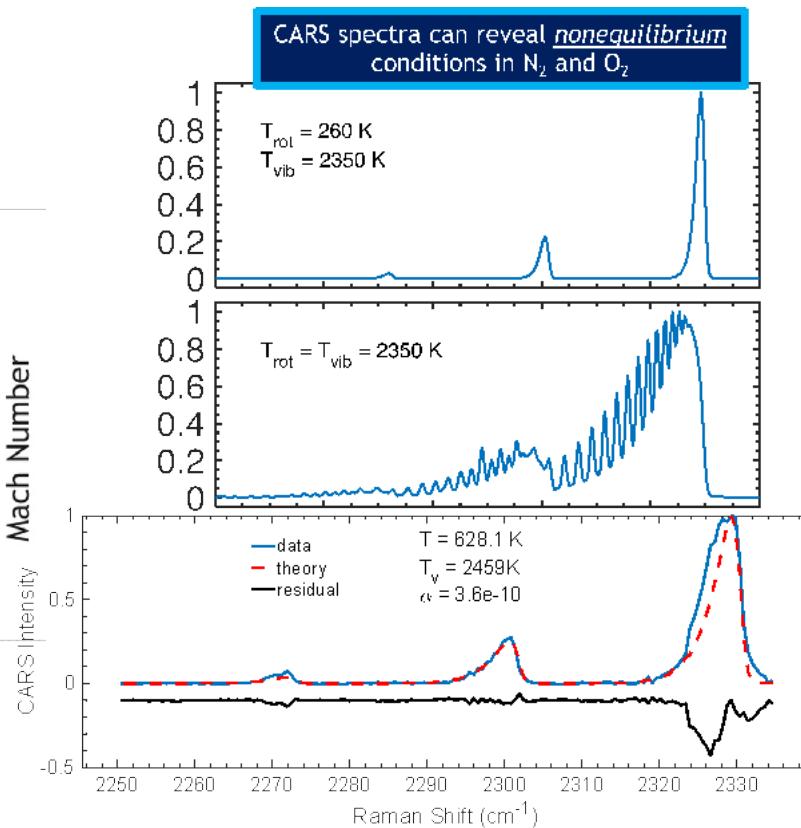
2X reduction in uncertainty!

- Short picosecond laser pulses enable high speed but result in noisy data
- We developed longer, nanosecond sources for high-speed CARS thermometry
- Demonstrated in flames
- Applied under nonequilibrium conditions in Sandia shock tunnel

## SPARC Calculation of Nozzle Nonequilibrium



New SPARC models  
guide experiment  
design



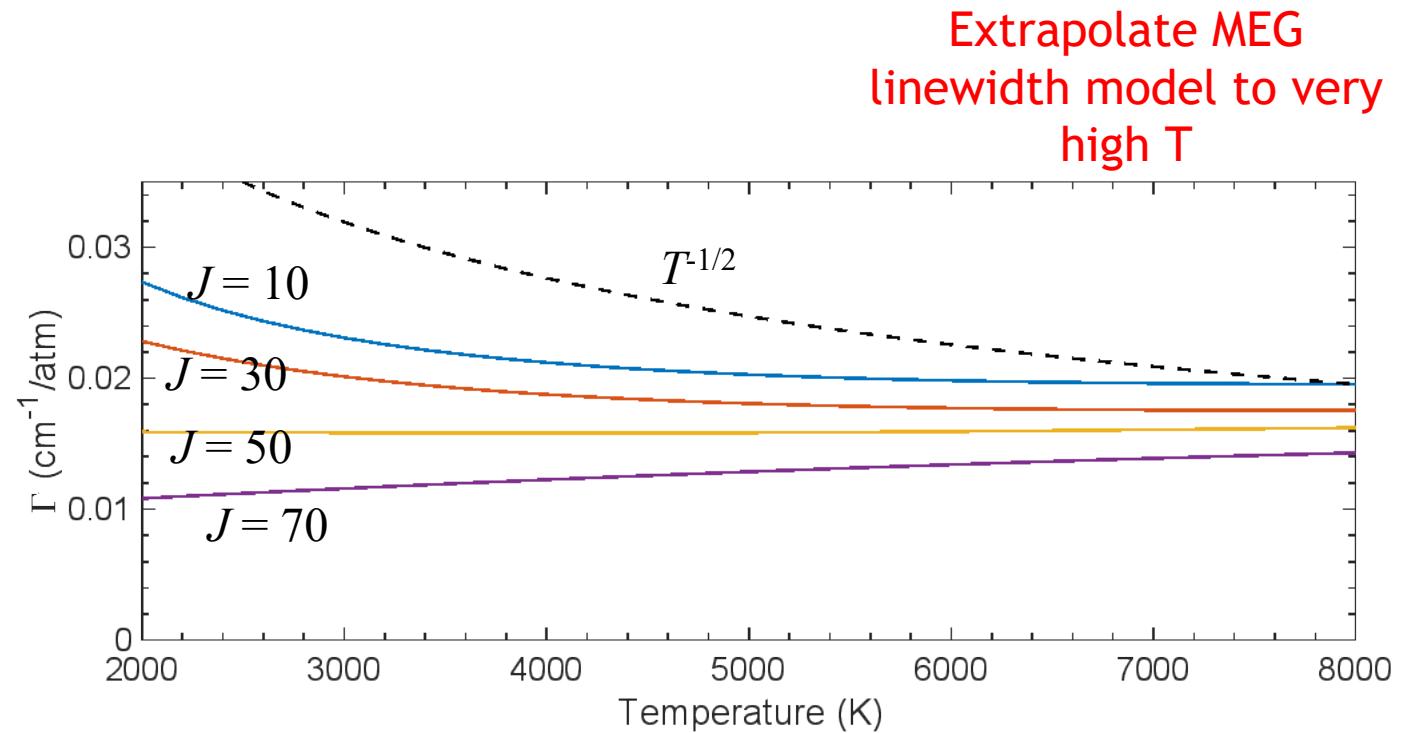
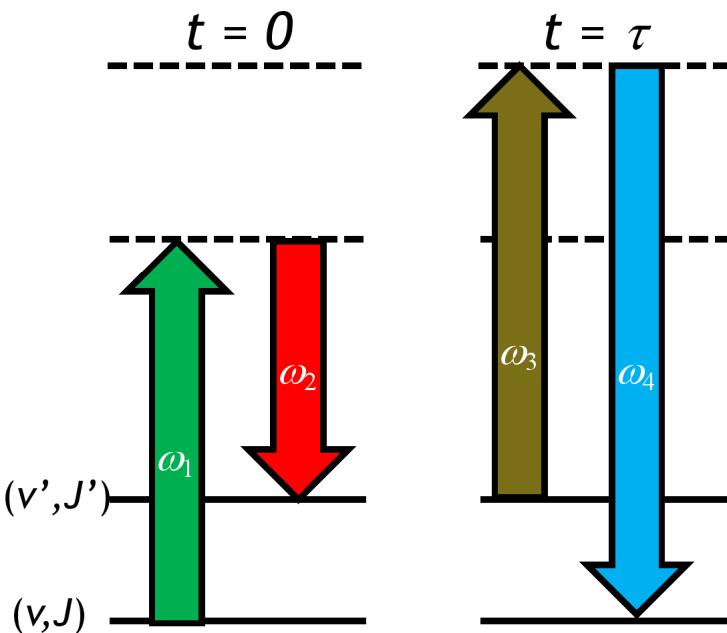
# Some Comments re: CARS Spectral Modeling



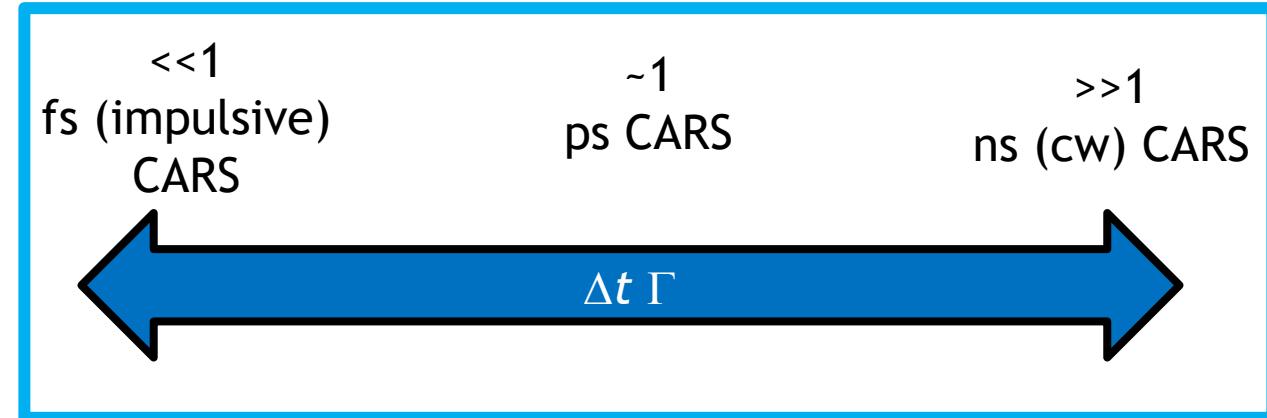
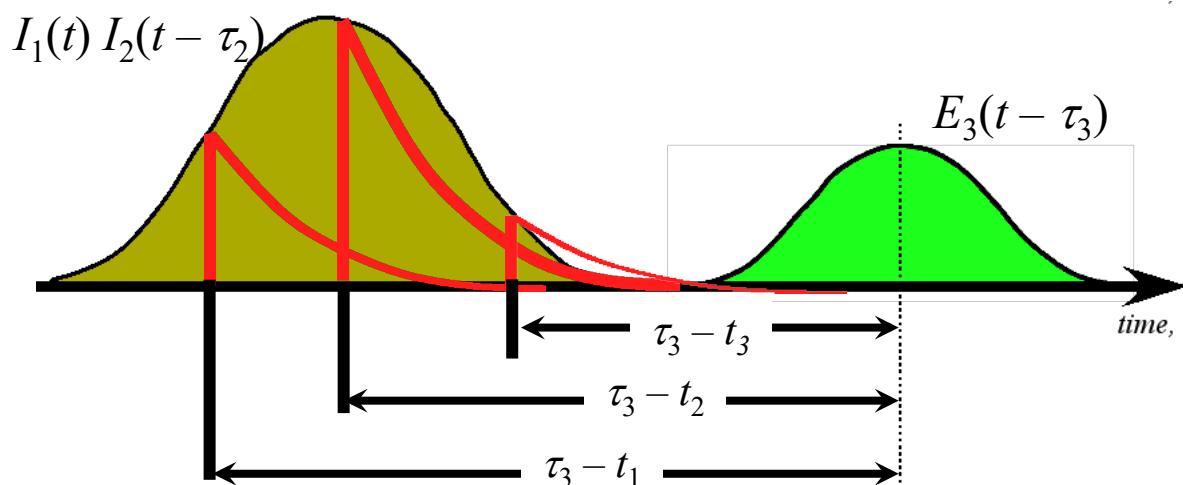
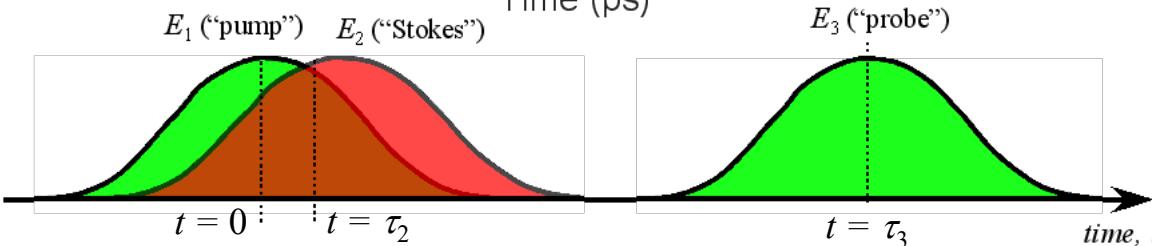
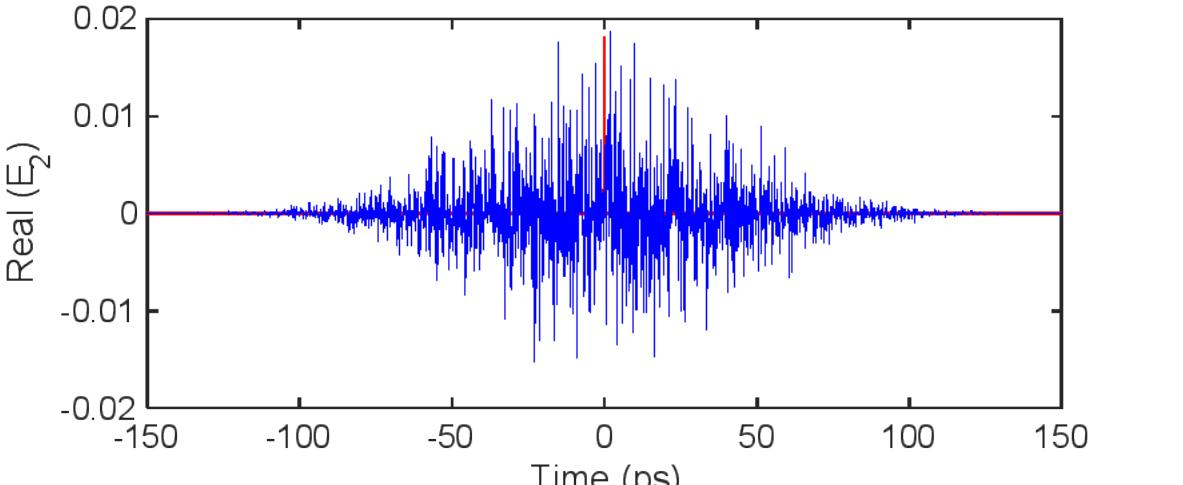
“Generalized Yuratich Equation,” Marrocco, *Opt. Lett.* **39**, 4831 (2014)

$$G(\omega; \tau) = \sum_{v,J} \Delta N_{v,J} \left( \frac{d\sigma}{d\Omega} \right)_{v,J} \int_{-\infty}^{\infty} \frac{E_3(\omega_3) e^{i\omega_3 \tau}}{[\omega_{v,J} - (\omega_3 - \omega)] - i\Gamma_J/2} d\omega_3$$

Phase factor  
(time delay)



## Picosecond CARS calculation



*Incoherent Sum of Impulsive Spectra*

$$\langle I_4(\omega; \tau_2, \tau_3) \rangle = \int_{-\infty}^{\infty} I_1(t) I_2(t - \tau_2) \|G(\omega; \tau_3 - t)\|^2 dt$$

$$G(\omega; \tau) = \sum_{v, J} \Delta N_{v, J} \left( \frac{d\sigma}{d\Omega} \right)_{v, J} \int_{-\infty}^{\infty} \frac{E_3(\omega_3) e^{i\omega_3 \tau}}{[\omega_{v, J} - (\omega_3 - \omega) - i\Gamma_J/2]} d\omega_3$$

# Summary and Conclusions



- CARS diagnostics are being applied multiple ground-test facilities to investigate physics of hypersonic flight
  - Extreme gas-phase temperatures (4000-6000 K)
  - Short-duration, impulsive experiments
- CARS thermometry of the  $\text{N}_2$  molecule appears to be effective at  $T$  as high as  $\sim 6000$  K
- Pulse-burst lasers can be adapted for CARS thermometry!
  - Picosecond pulses are enabling but noisy
  - Nanosecond pulses can reduce CARS noise
  - Short pulses provide superior performance but not yet ready for burst-mode application (Purdue/SE!)
- We have rigorously developed a new method for treating picosecond CARS spectra

