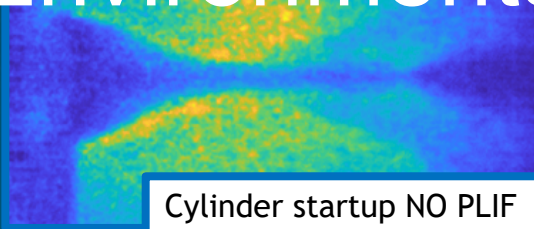
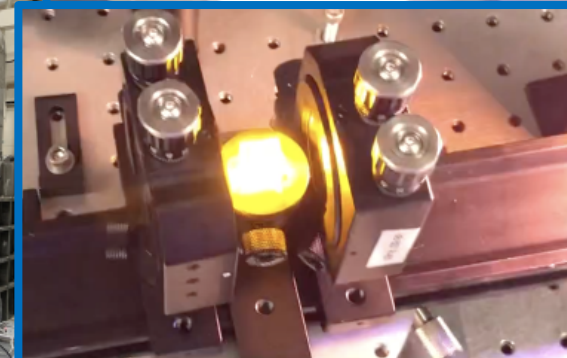
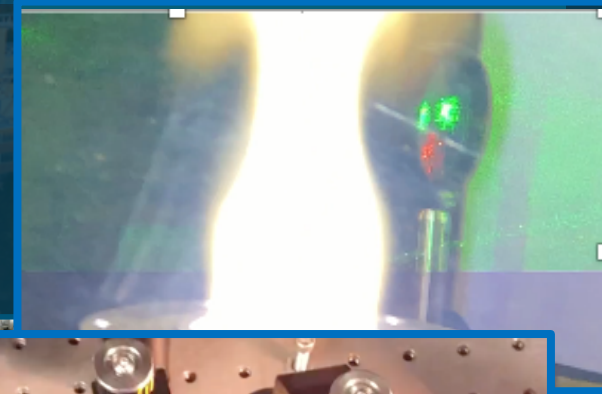
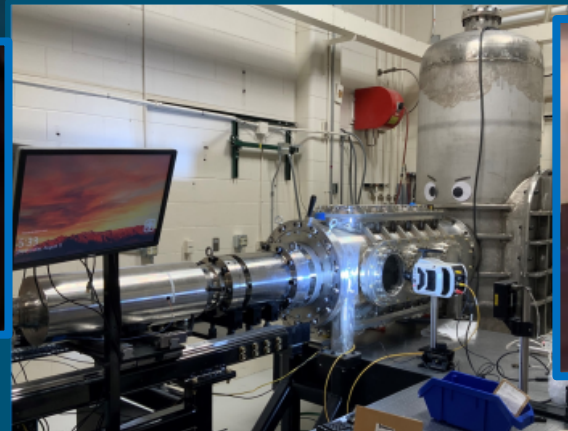




# CARS Thermometry and Species Measurements in Extreme-Temperature Environments



Cylinder startup NO PLIF



Sean P. Kearney  
Engineering Sciences Center  
Sandia National Laboratories  
Albuquerque, NM 87175  
[spkearn@sandia.gov](mailto:spkearn@sandia.gov)

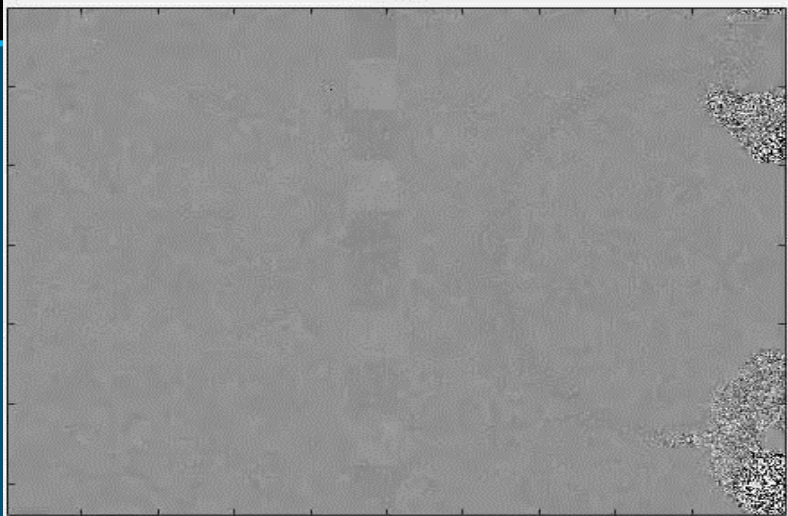


Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

## 2 Our Motivation: High Temperature Materials Test Environments

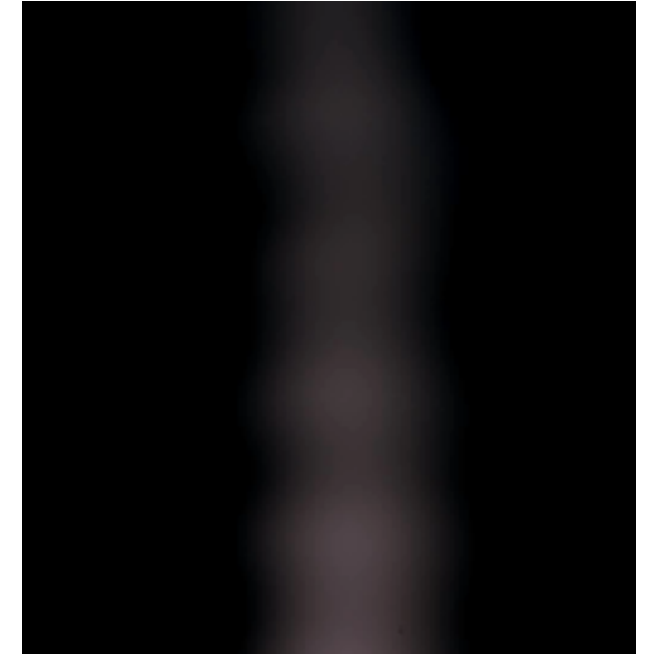
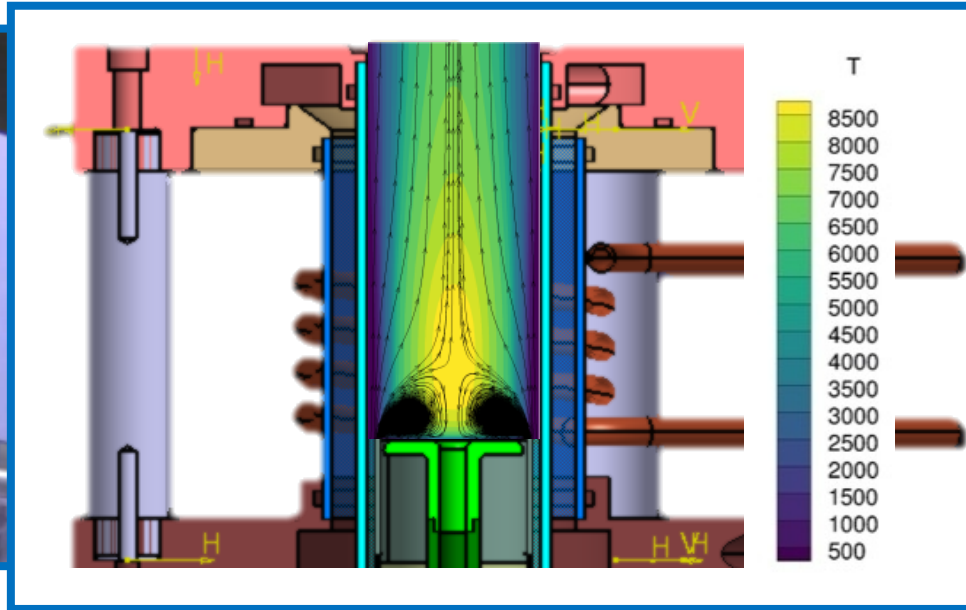
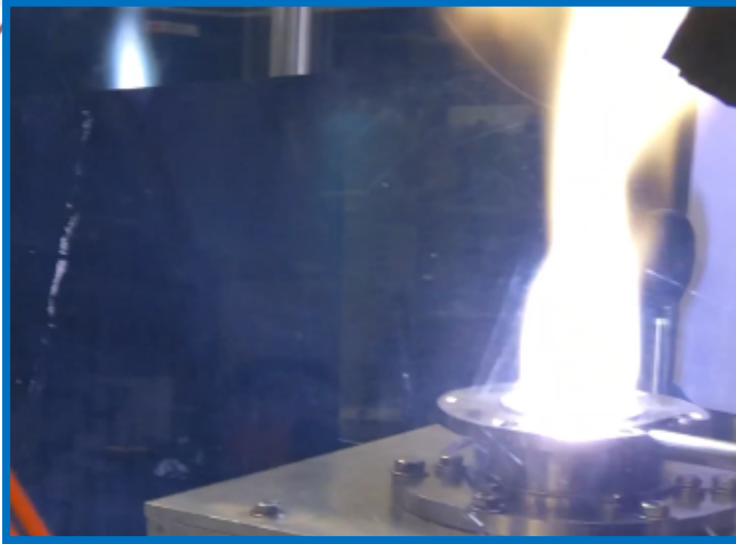


### Free-Piston Reflected Shock Tunnel



Inductively Coupled Plasma Torch

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



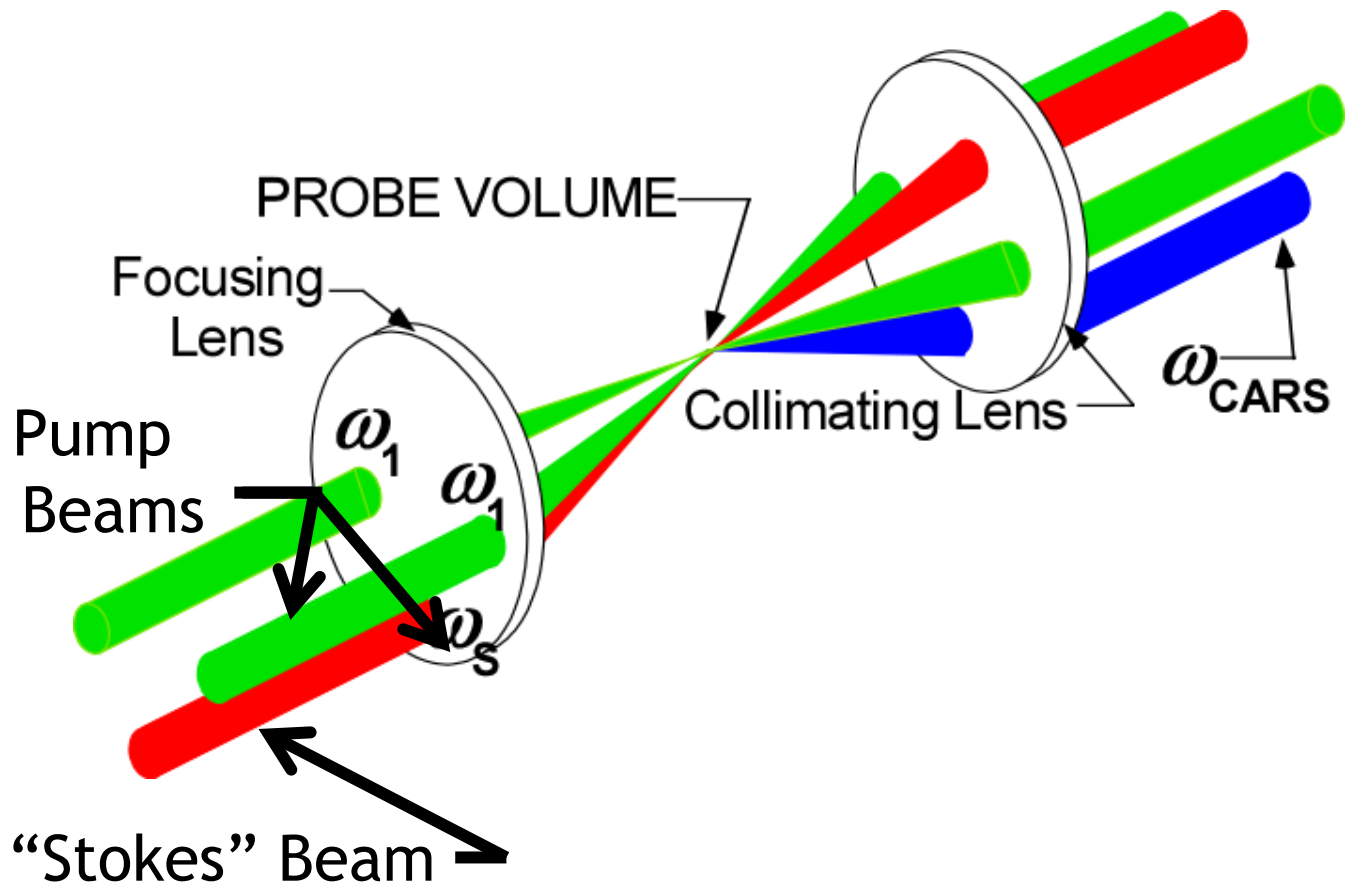
## Inductively Coupled Plasma Torch

- 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

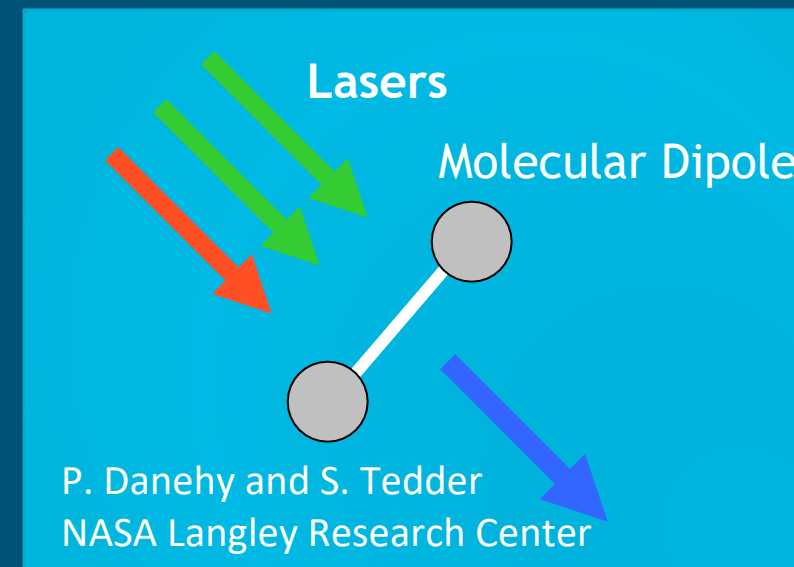
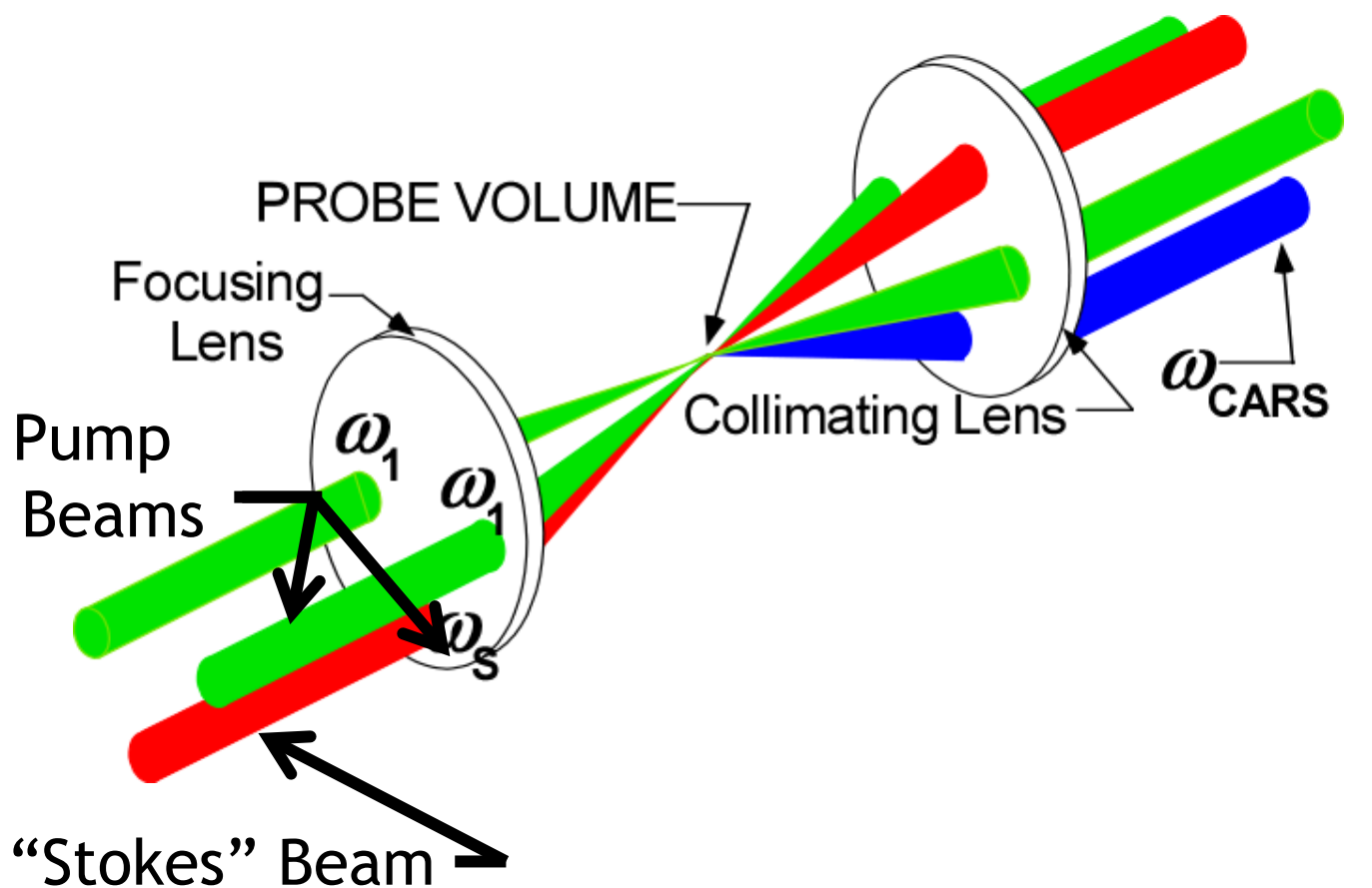


# Coherent anti-Stokes Raman scattering (CARS)

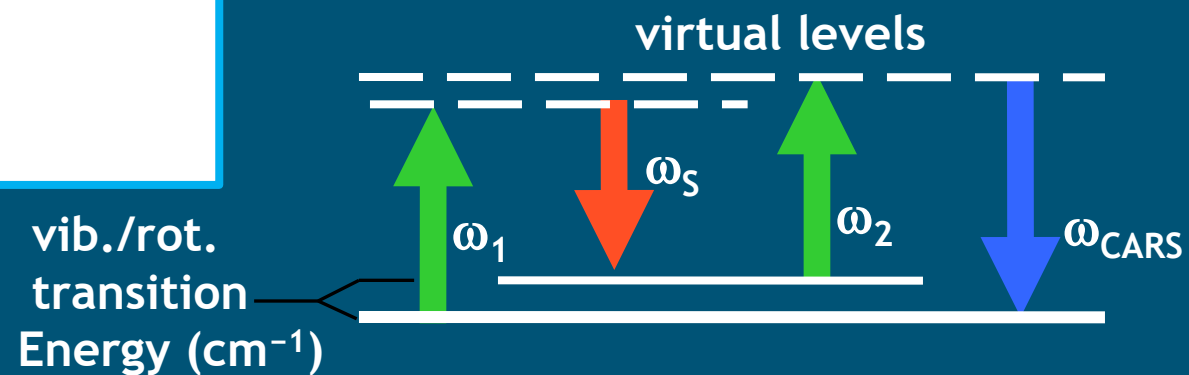




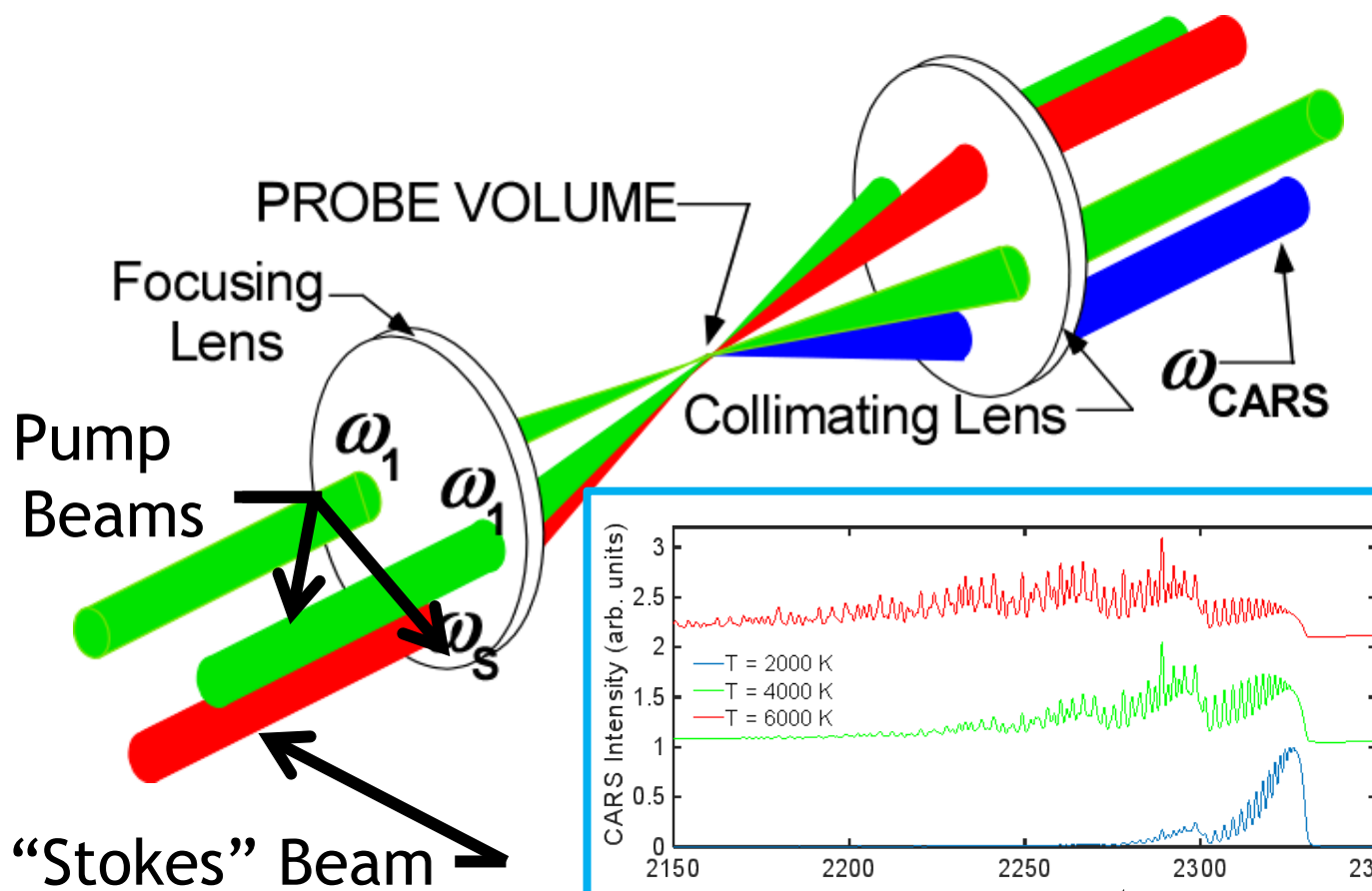
# Coherent anti-Stokes Raman scattering (CARS)



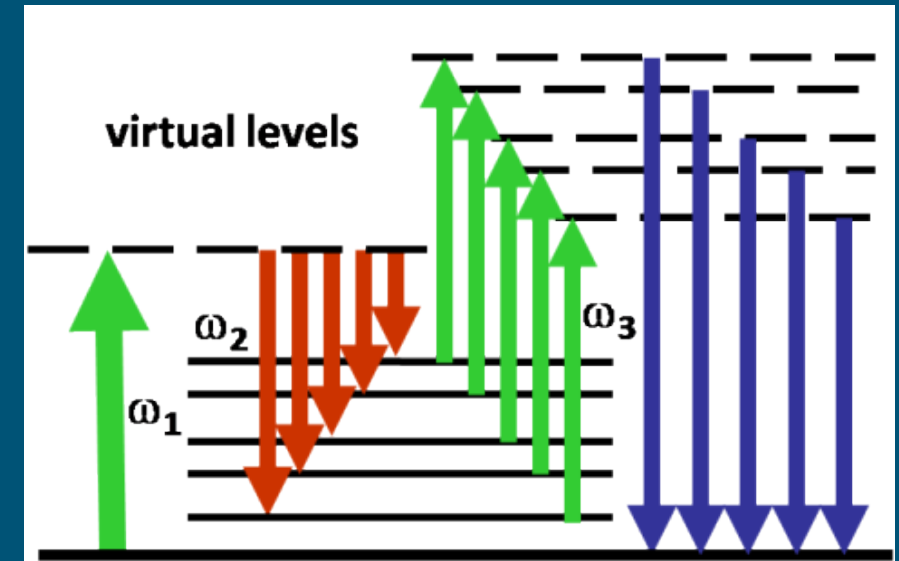
Coherent Anti-Stokes Raman



# Coherent anti-Stokes Raman scattering (CARS)



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



Broadband Stokes Source Pumps Multiple Raman transitions simultaneously



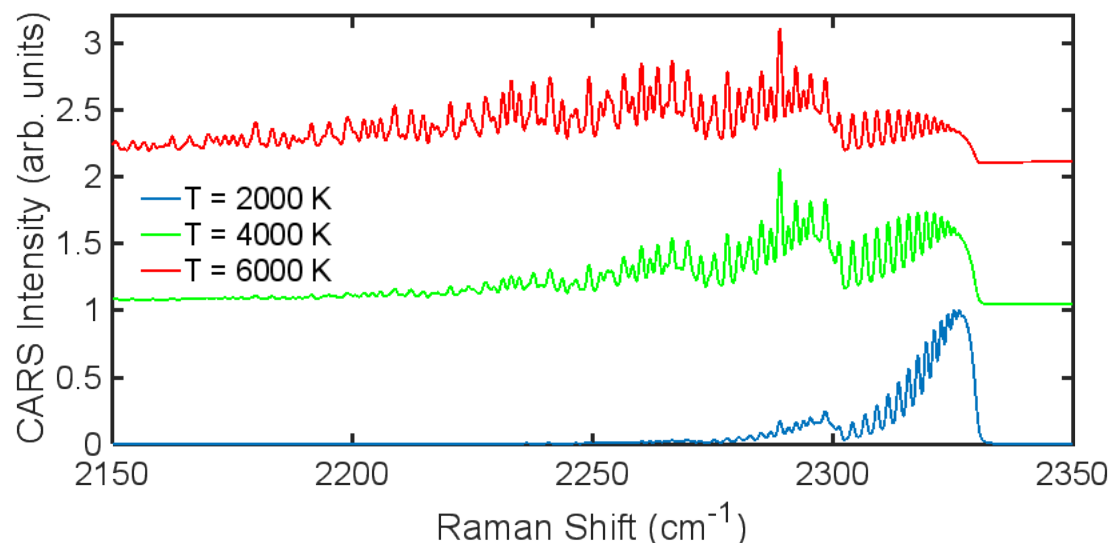
# IC Plasma Torch N<sub>2</sub> Thermometry

## "Conventional" CARS Instrument

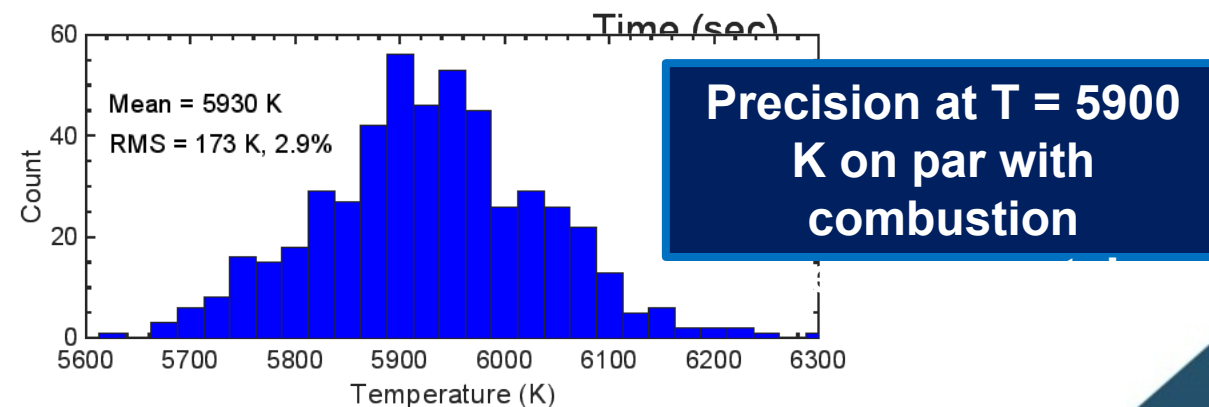
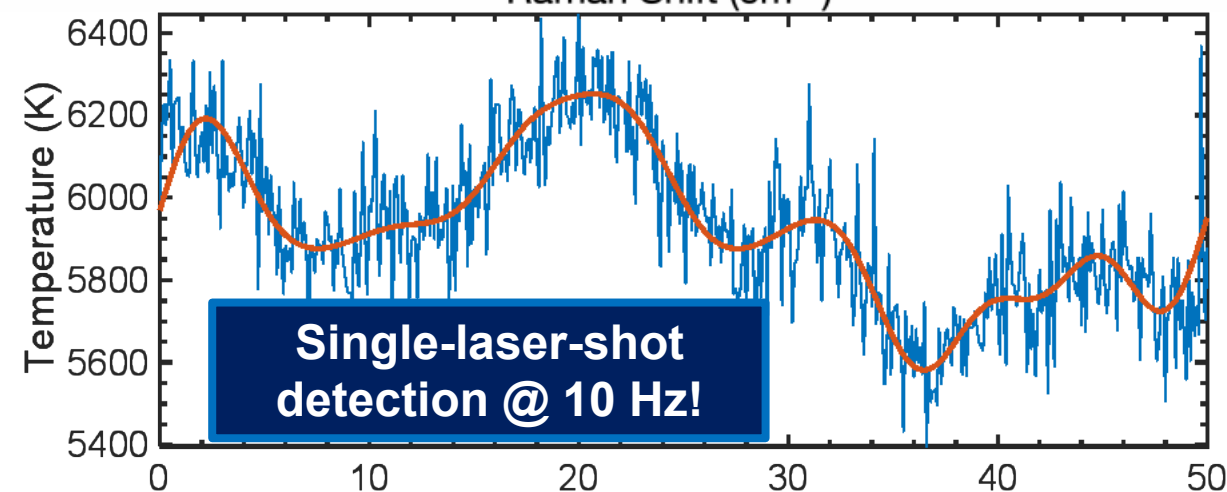
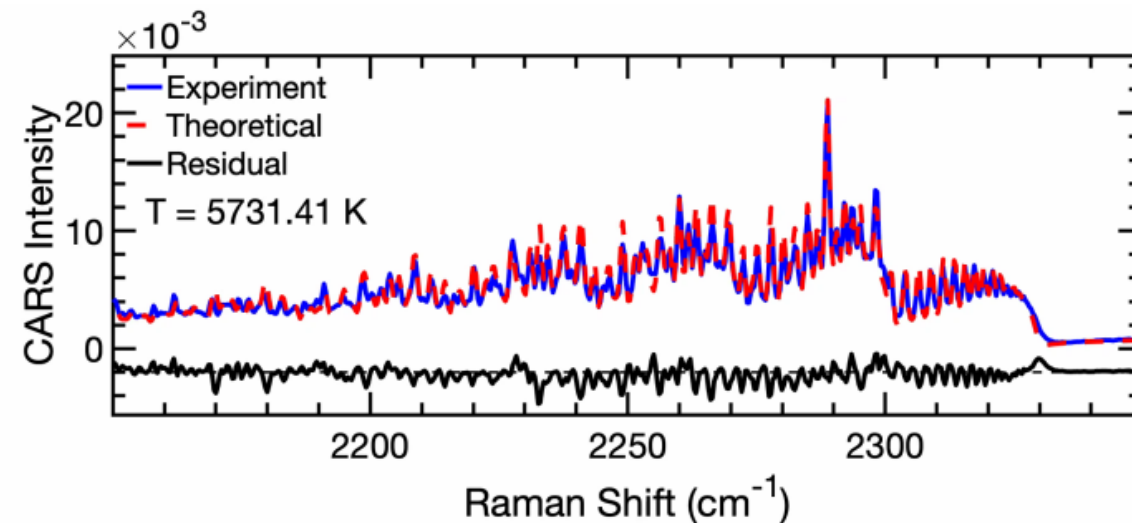
- ~10-ns laser pulses at 10-Hz
- Broadband dye laser for Stokes source

## Key technical challenges

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

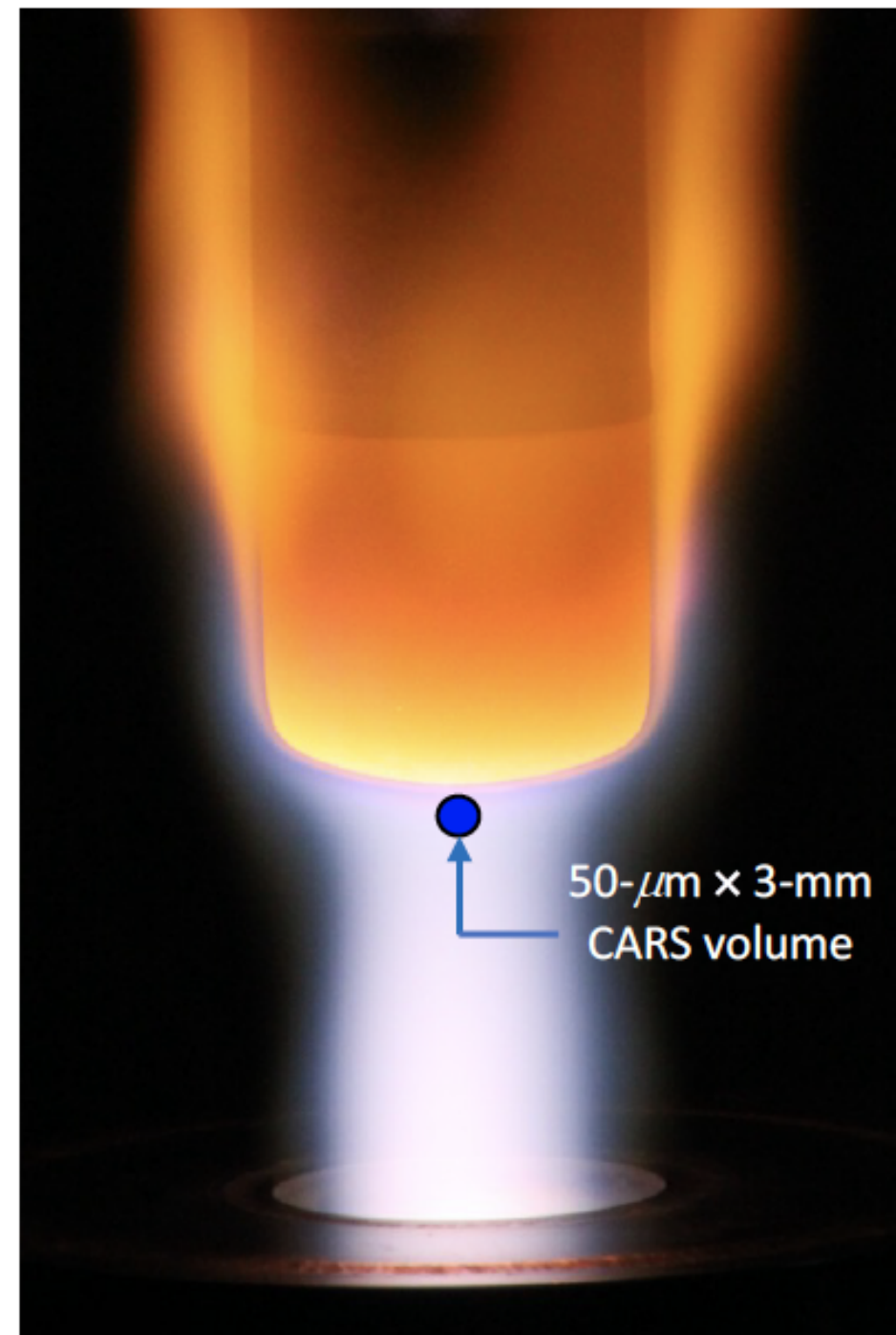
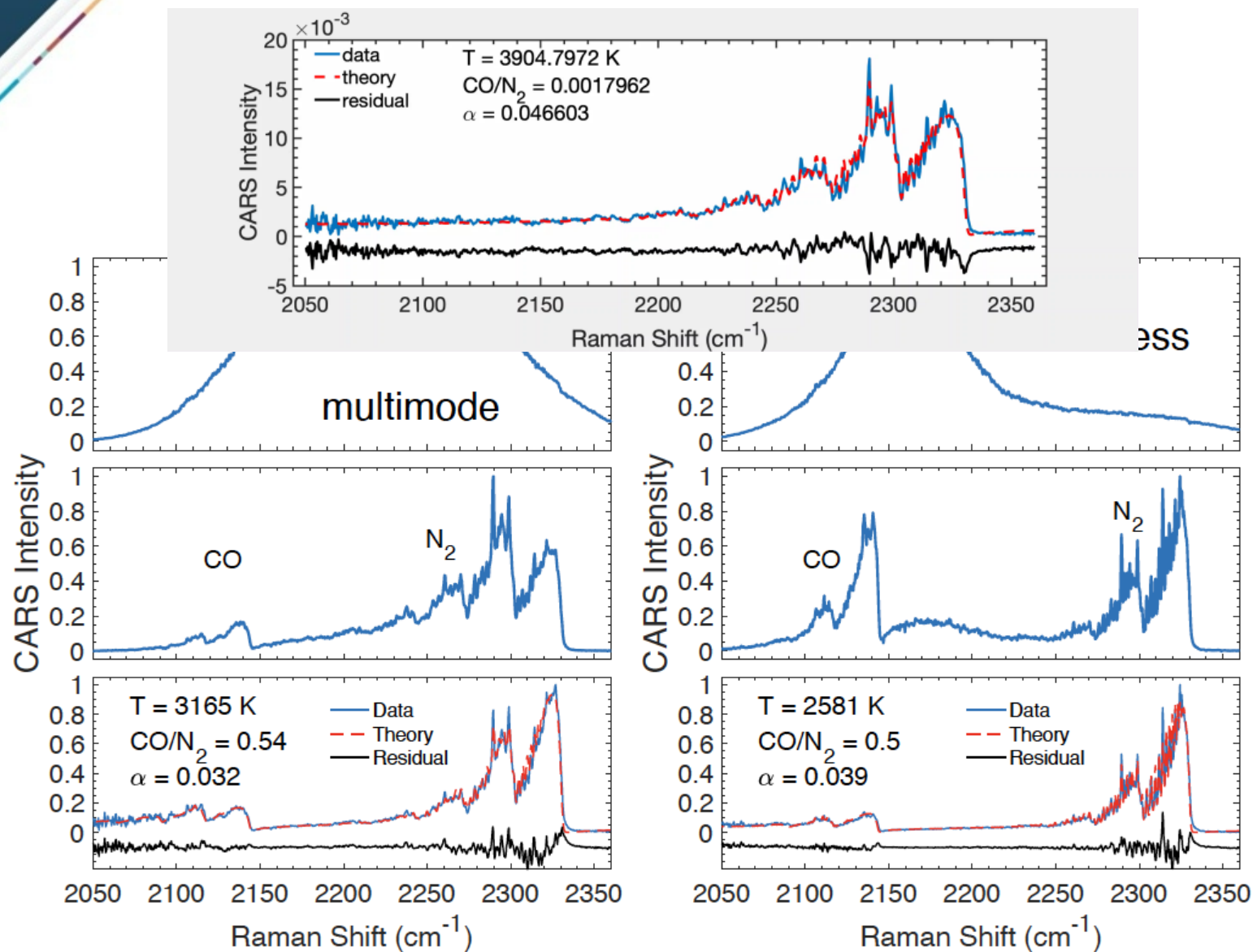


Calculated N<sub>2</sub> CARS Spectra





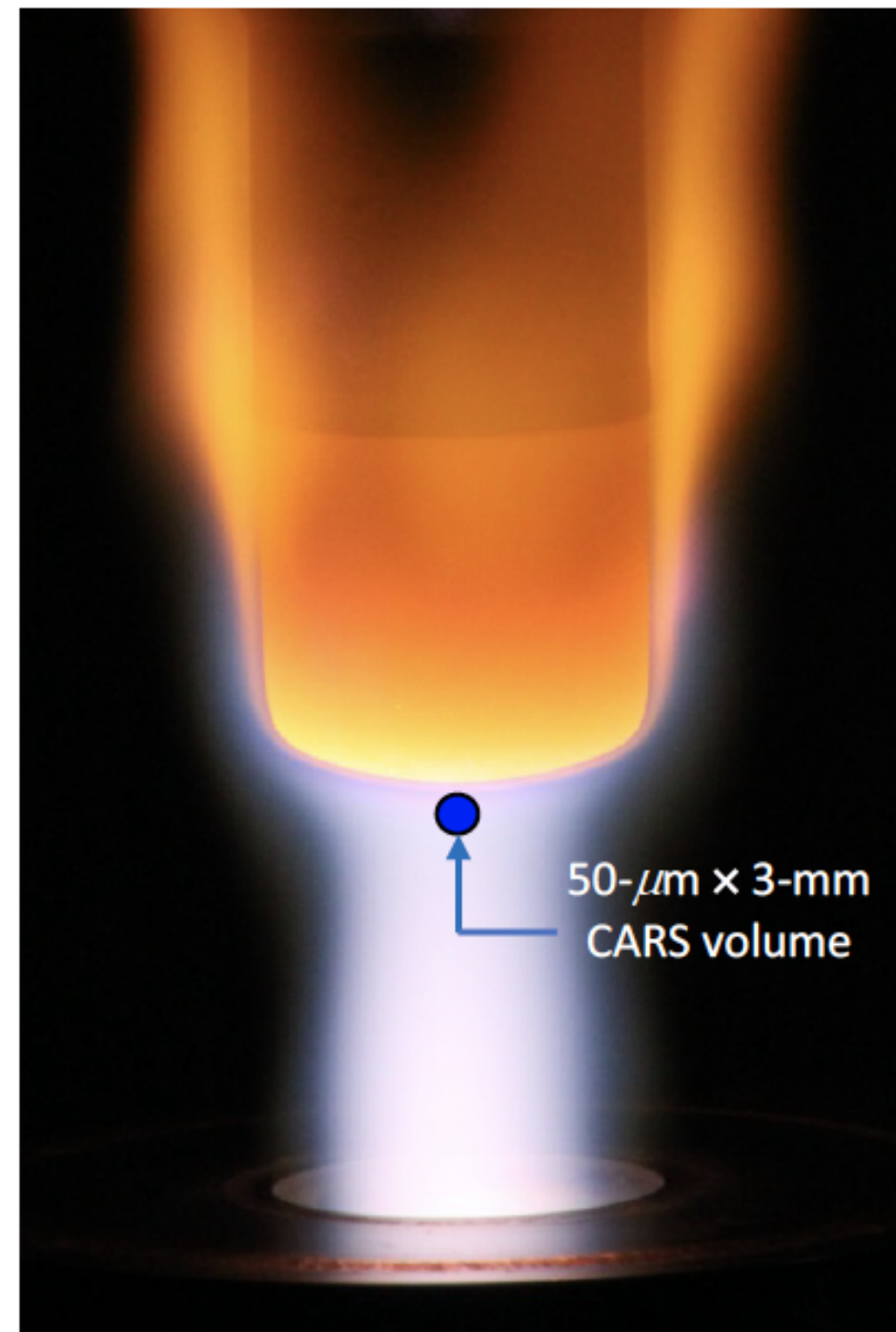
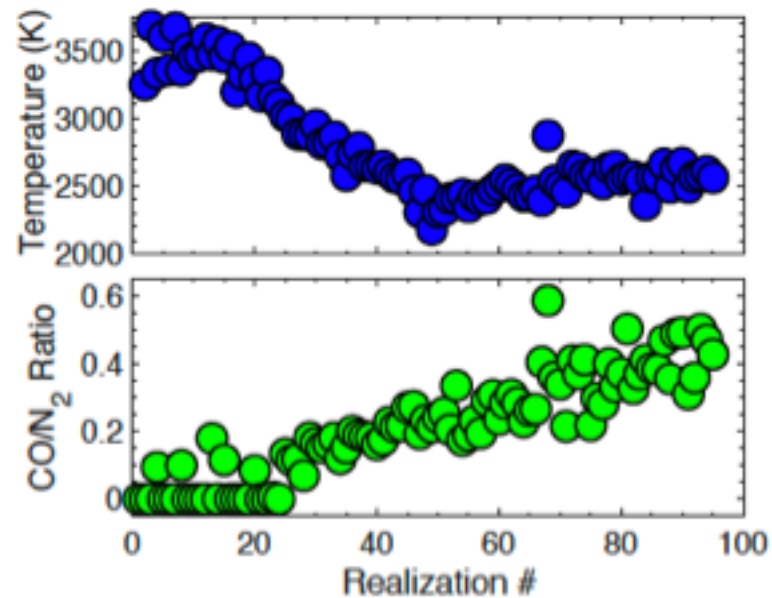
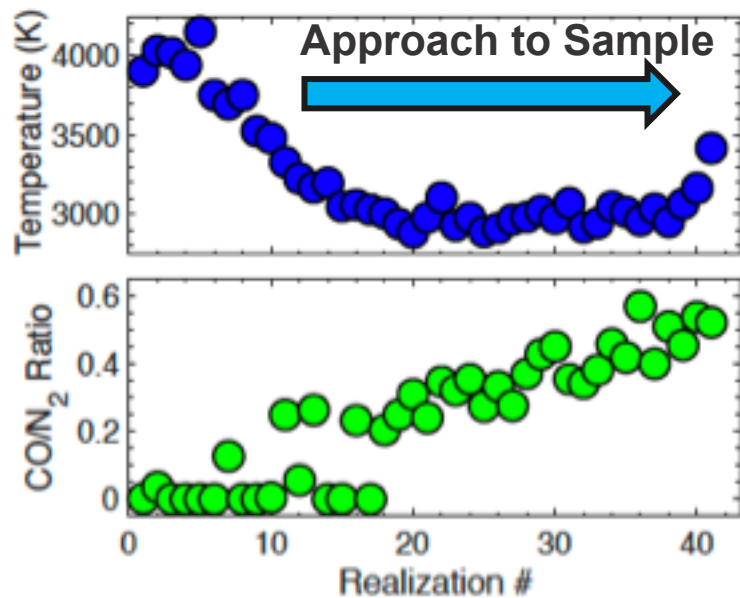
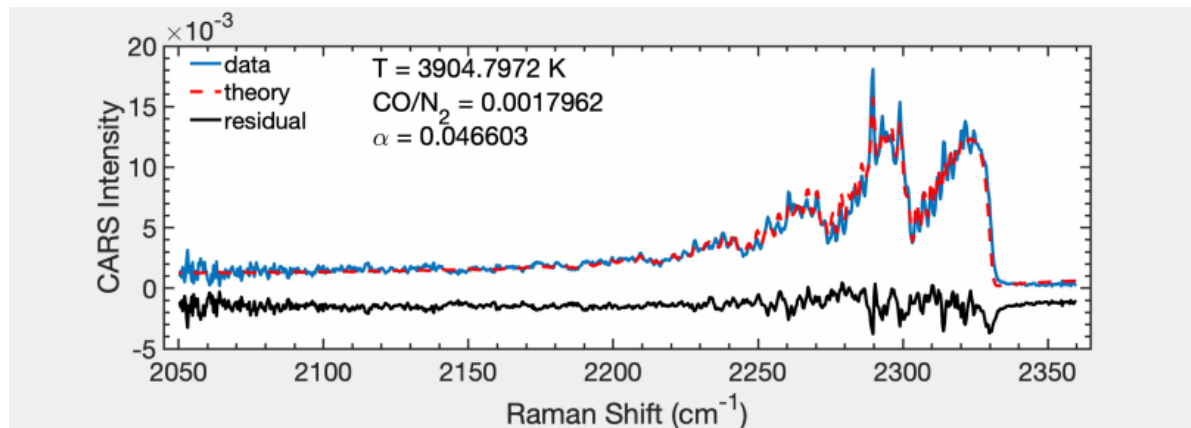
# CO/Temperature Measurements in High-T Reaction Layer



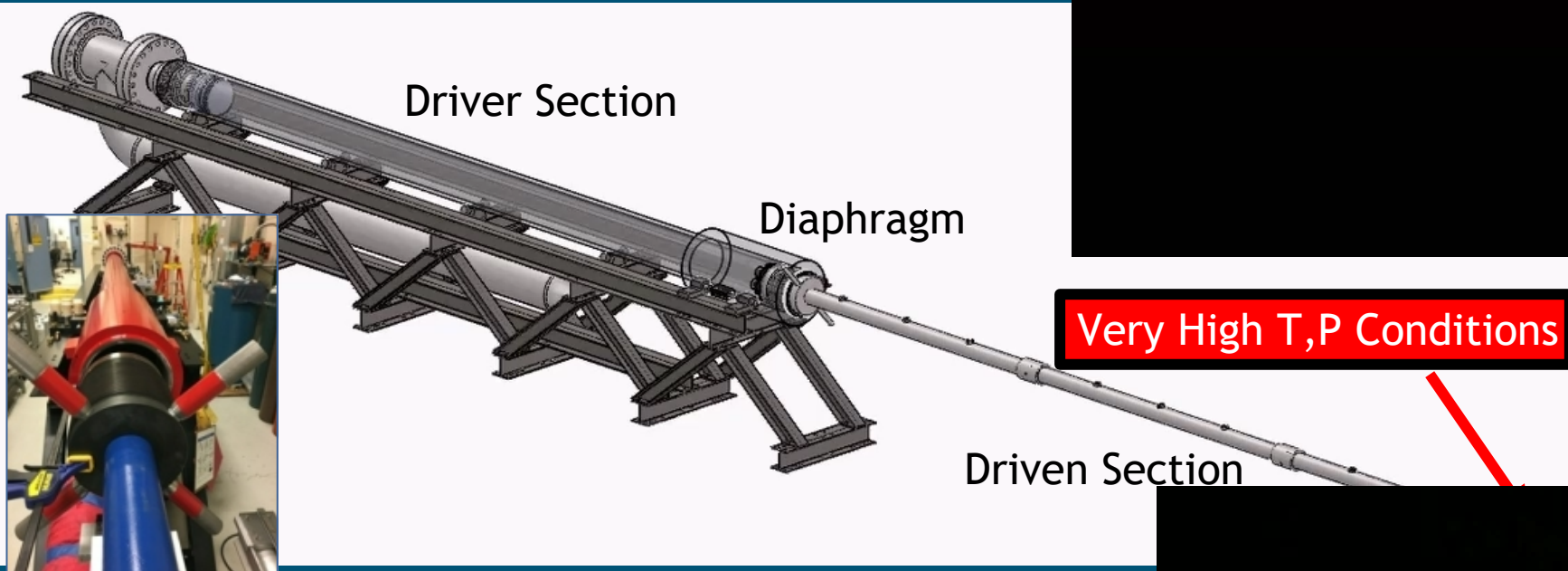




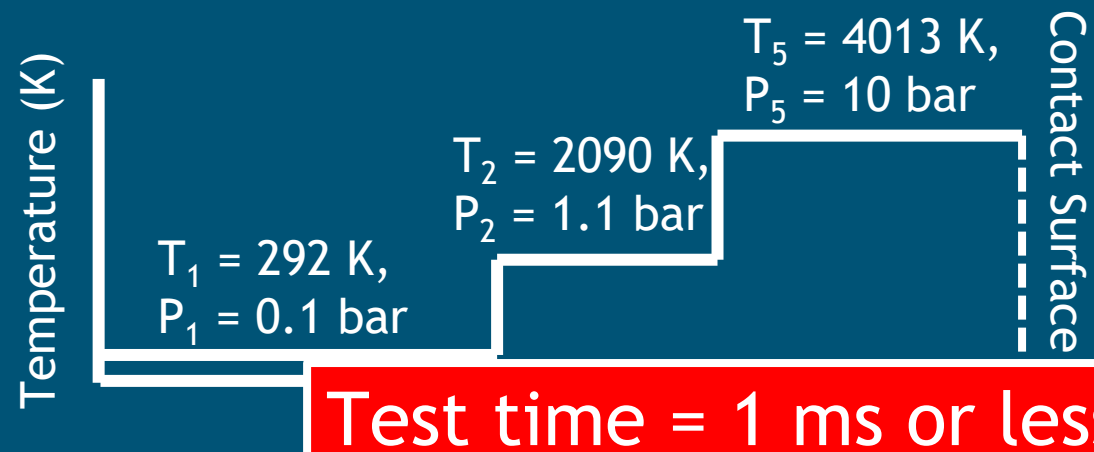
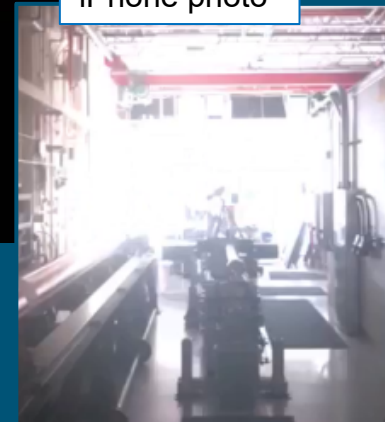
# CO/Temperature Measurements in High-T Reaction Layer



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

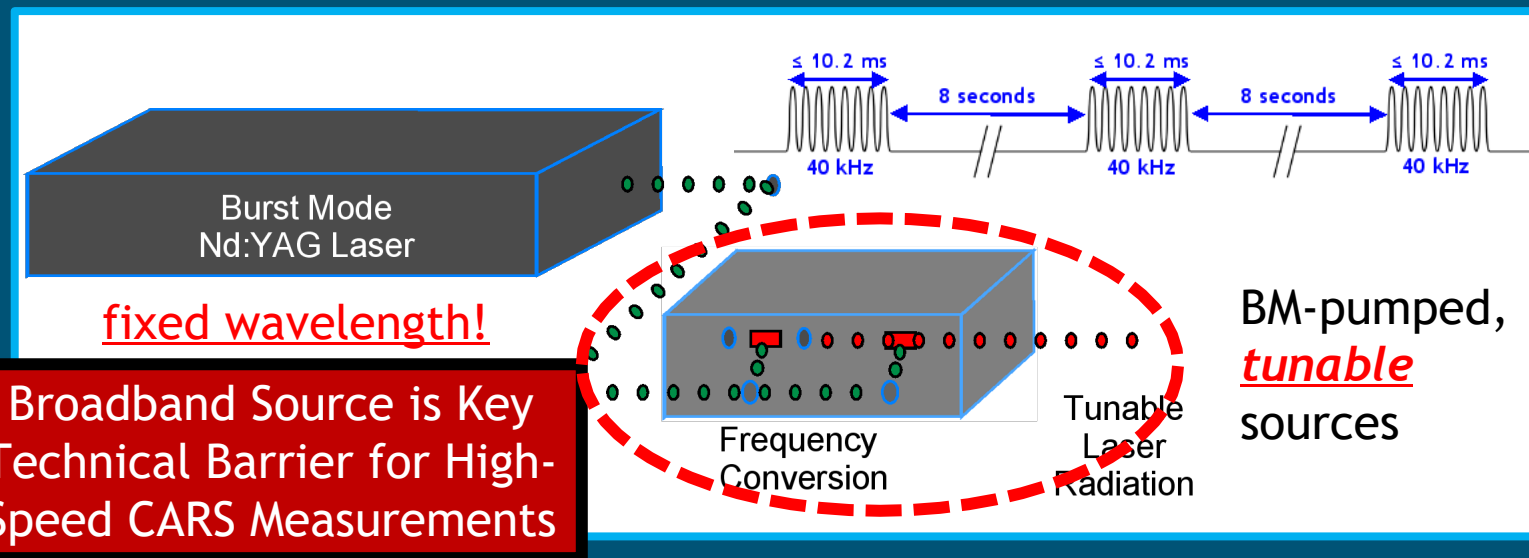


iPhone photo



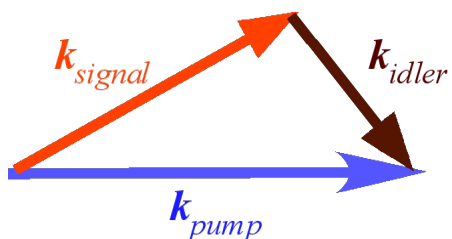
# Picosecond Pulse-Burst CARS at 100 kHz for HST Application

- 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)

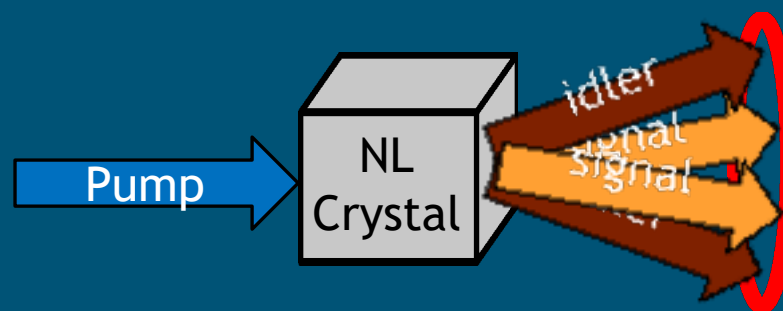
### Phase Matching Requirements



$$k_{pump} = k_{signal} + k_{idler}$$

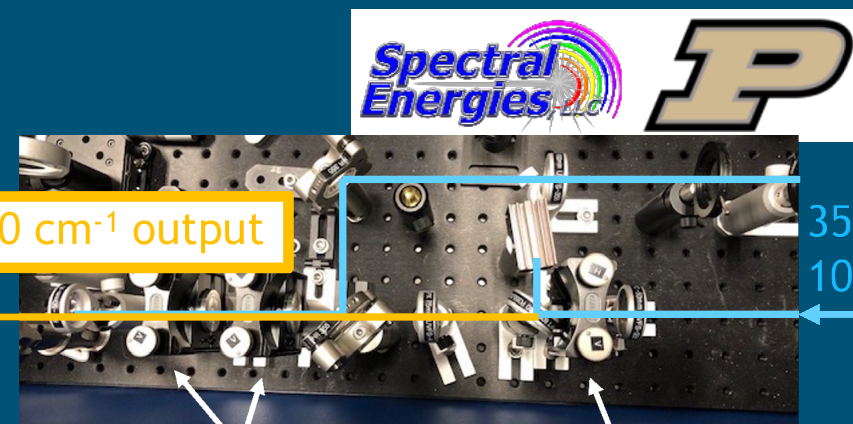
$$\omega_{pump} = \omega_{signal} + \omega_{idler}$$

$$|k| = 2\pi/\lambda, \omega = 1/\lambda$$



Picosecond OPG = Enabling Technology!

100-120 cm<sup>-1</sup> output

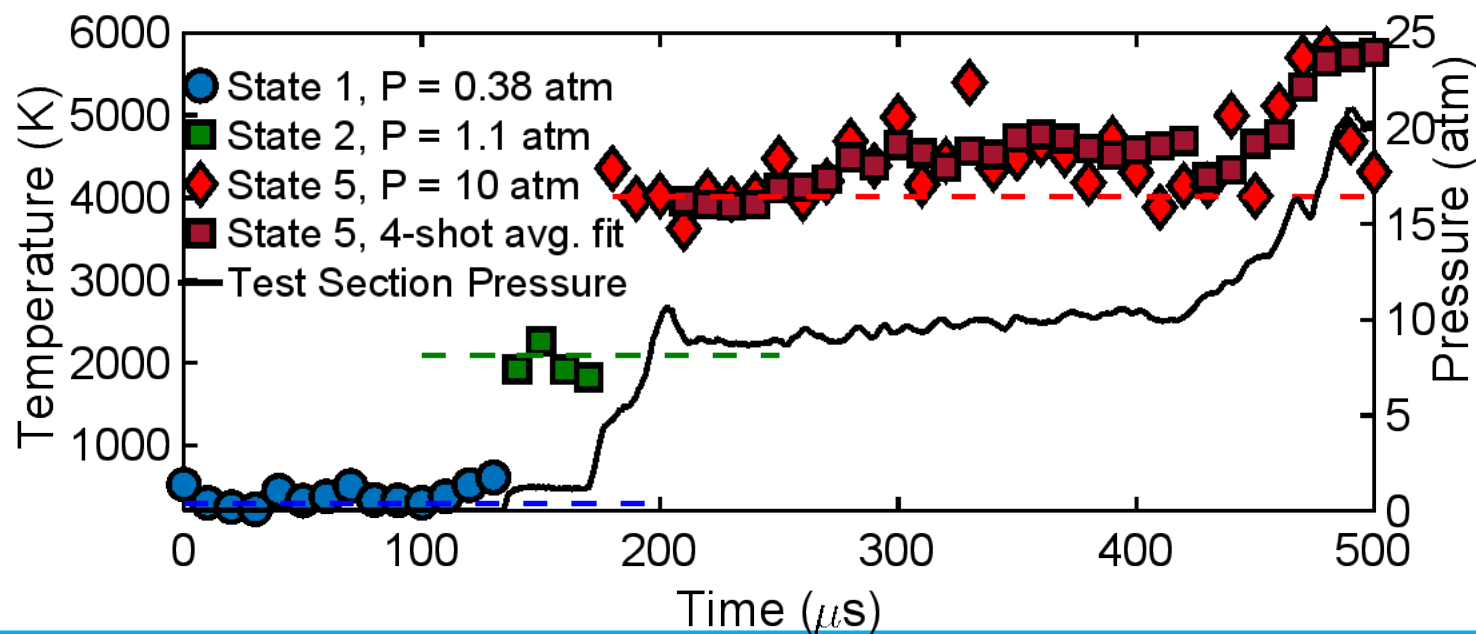
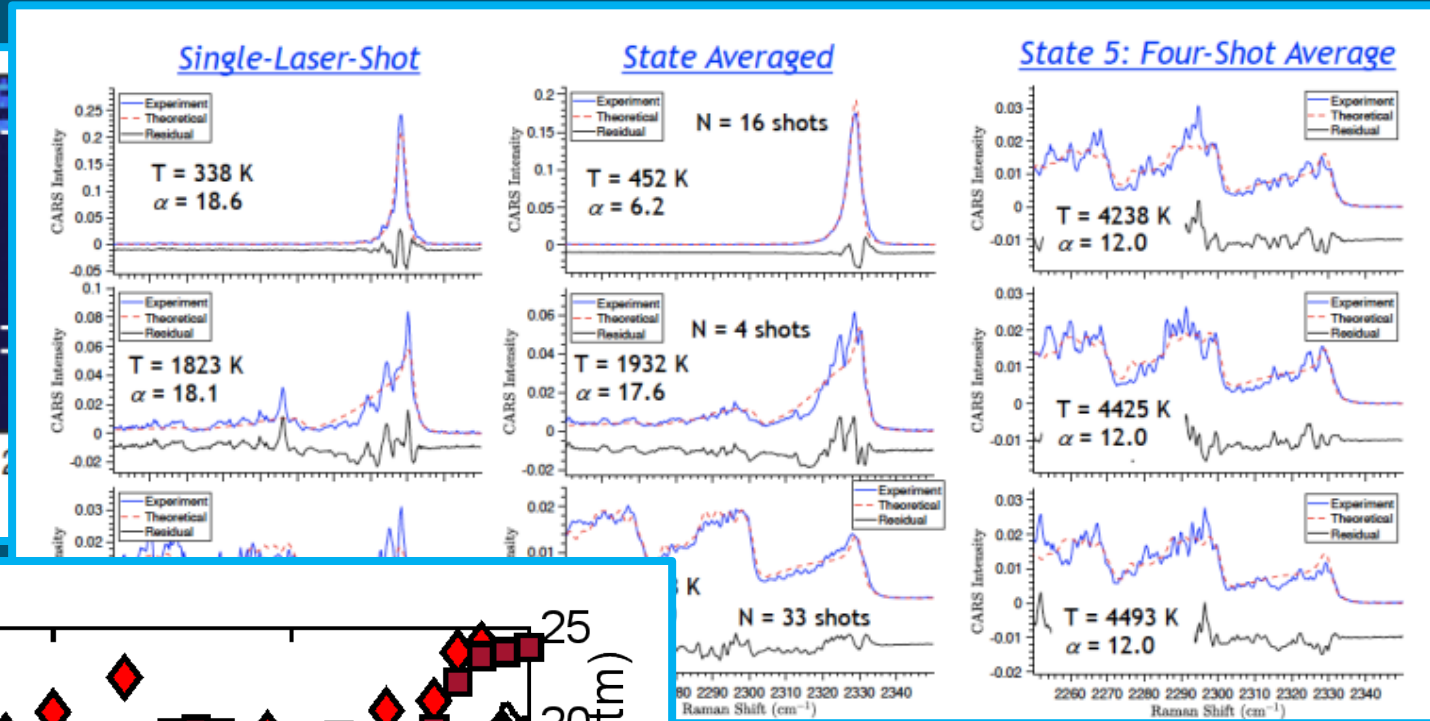
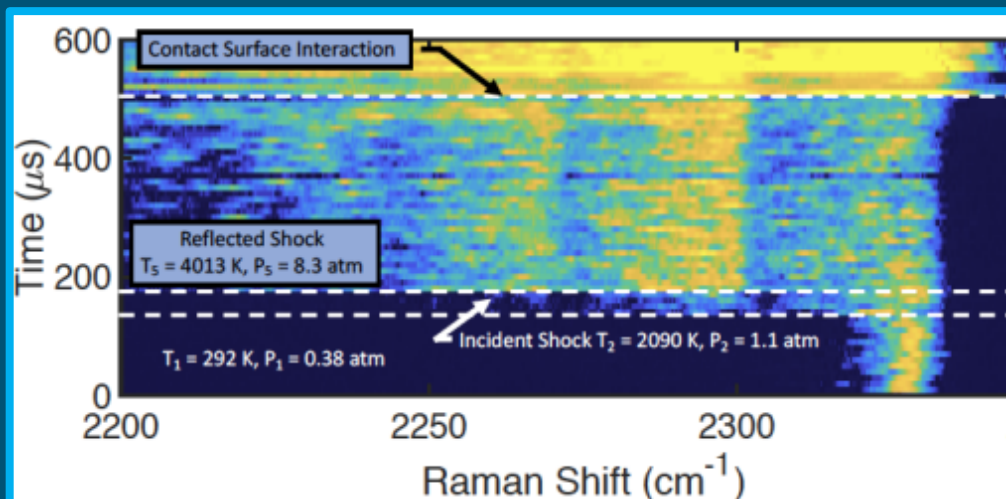


OPA section

OPG crystal



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



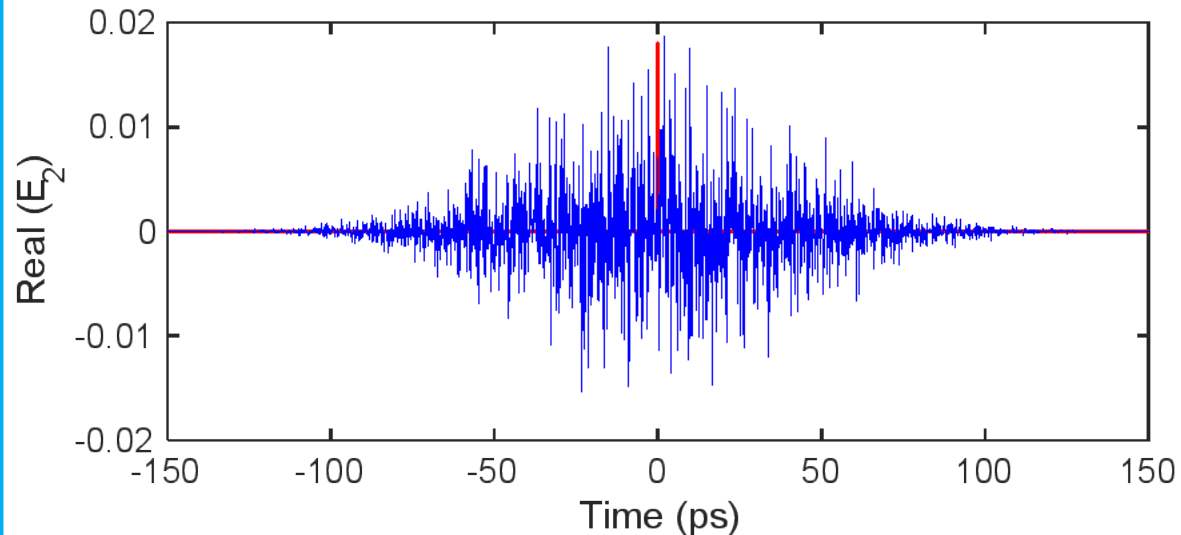


# Picosecond CARS comes with a single-shot noise penalty

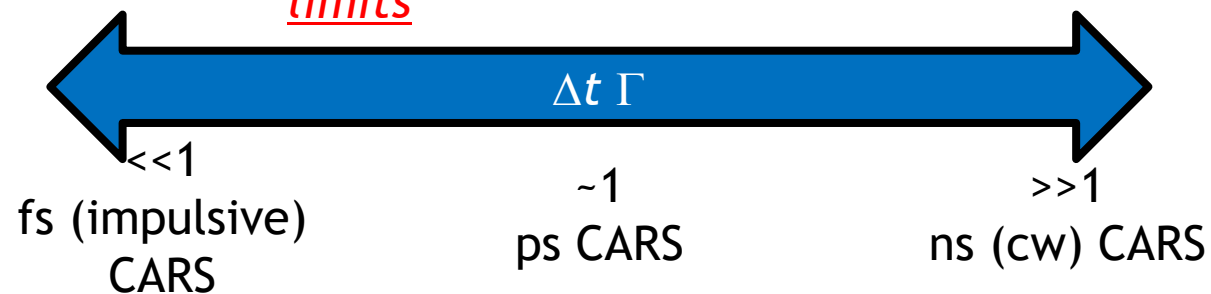
Dominant source of CARS noise is the quality of the broadband pulse

Time-Bandwidth Product - Fourier-Transform Limit

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



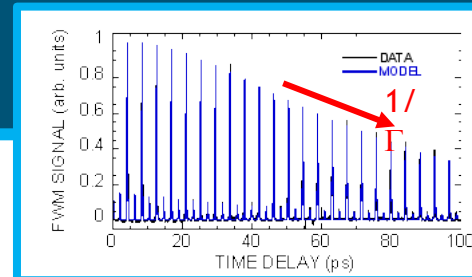
Picosecond CARS lies between cw and impulsive limits



Transform limited  
(low noise)

Time averaged

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



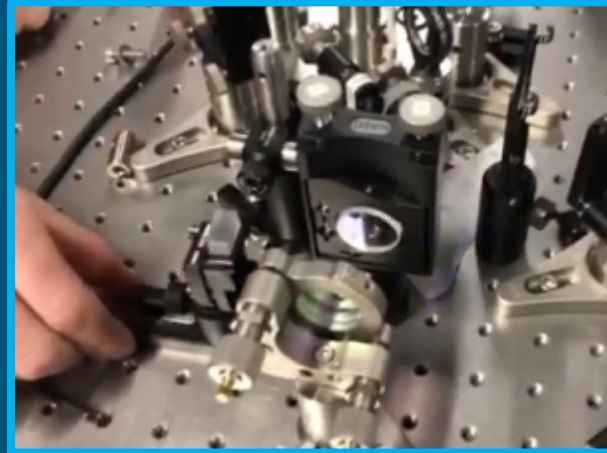
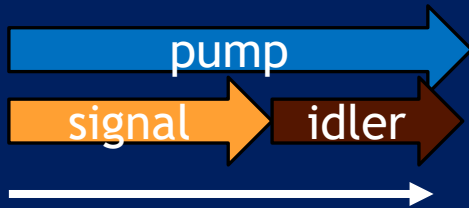
• To achieve sufficient bandwidth for CARS, pulse must exhibit  $\sim 150\text{-fs}$  features - inherently noisy!

# Nanosecond Burst-Mode CARS via Noncolinear Optical Parametric Oscillator



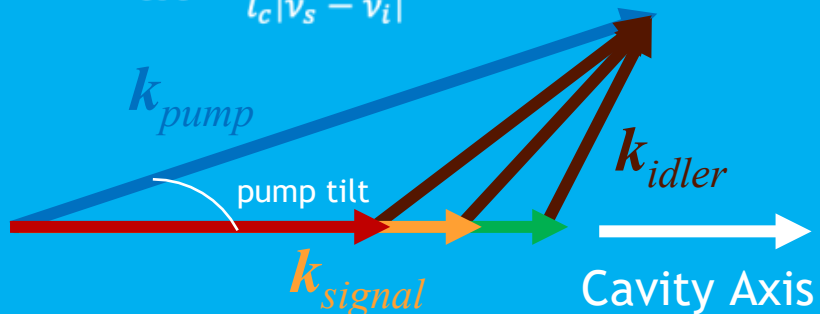
Conversion of low-intensity nanosecond pulses requires laser-cavity gain

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



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

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

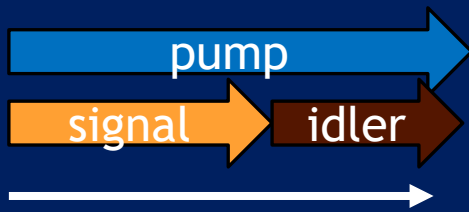


# Nanosecond Burst-Mode CARS via Noncolinear Optical Parametric Oscillator



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

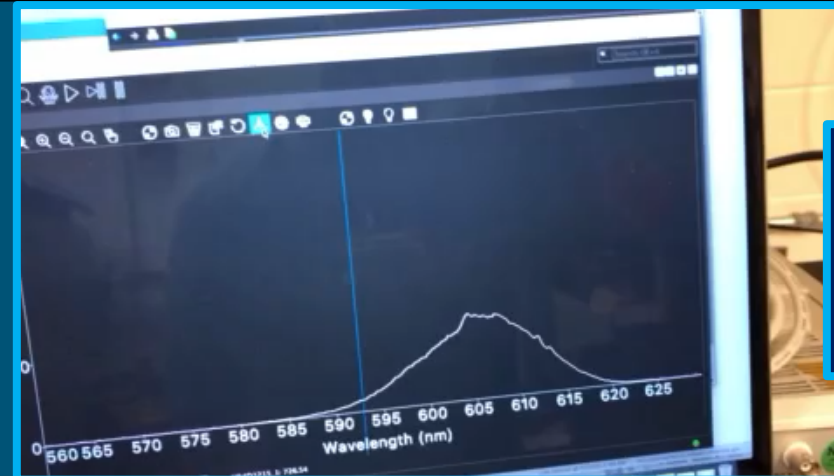
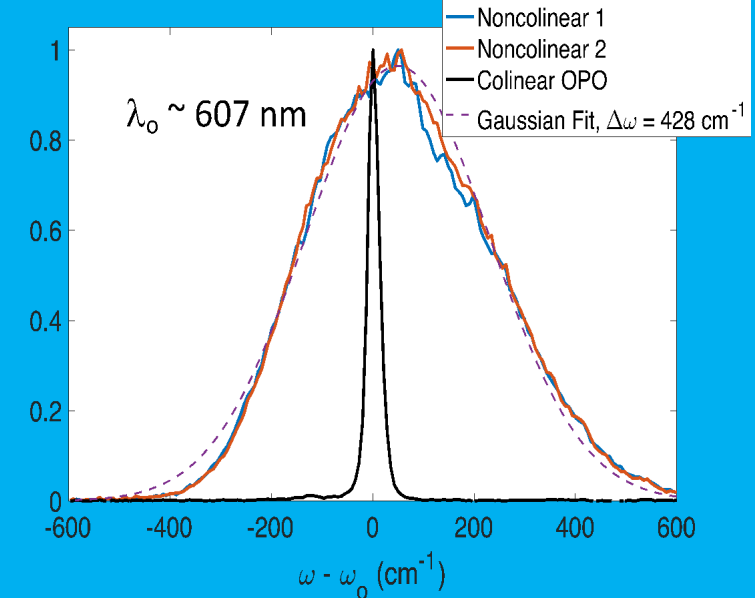
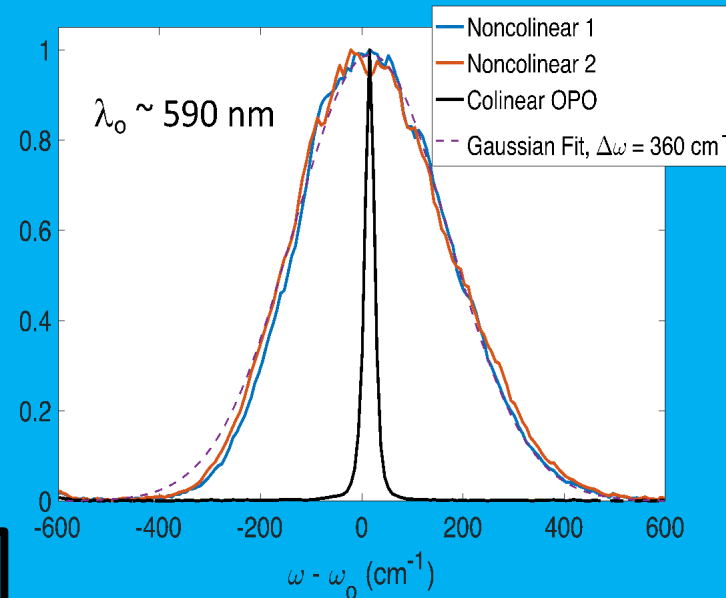
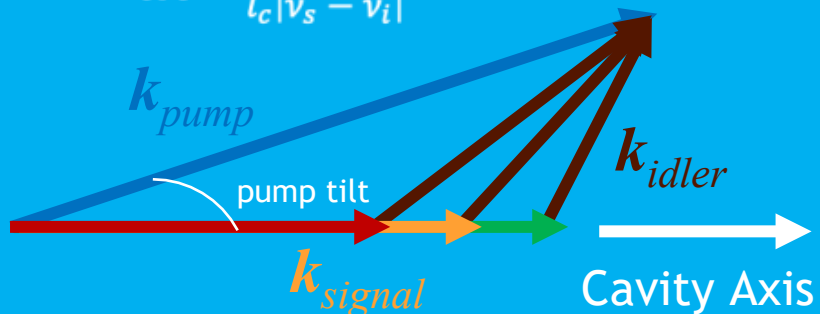
Most OPOs are co-linear to satisfy phase-matching constraint



constrained to  
laser cavity axis

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

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

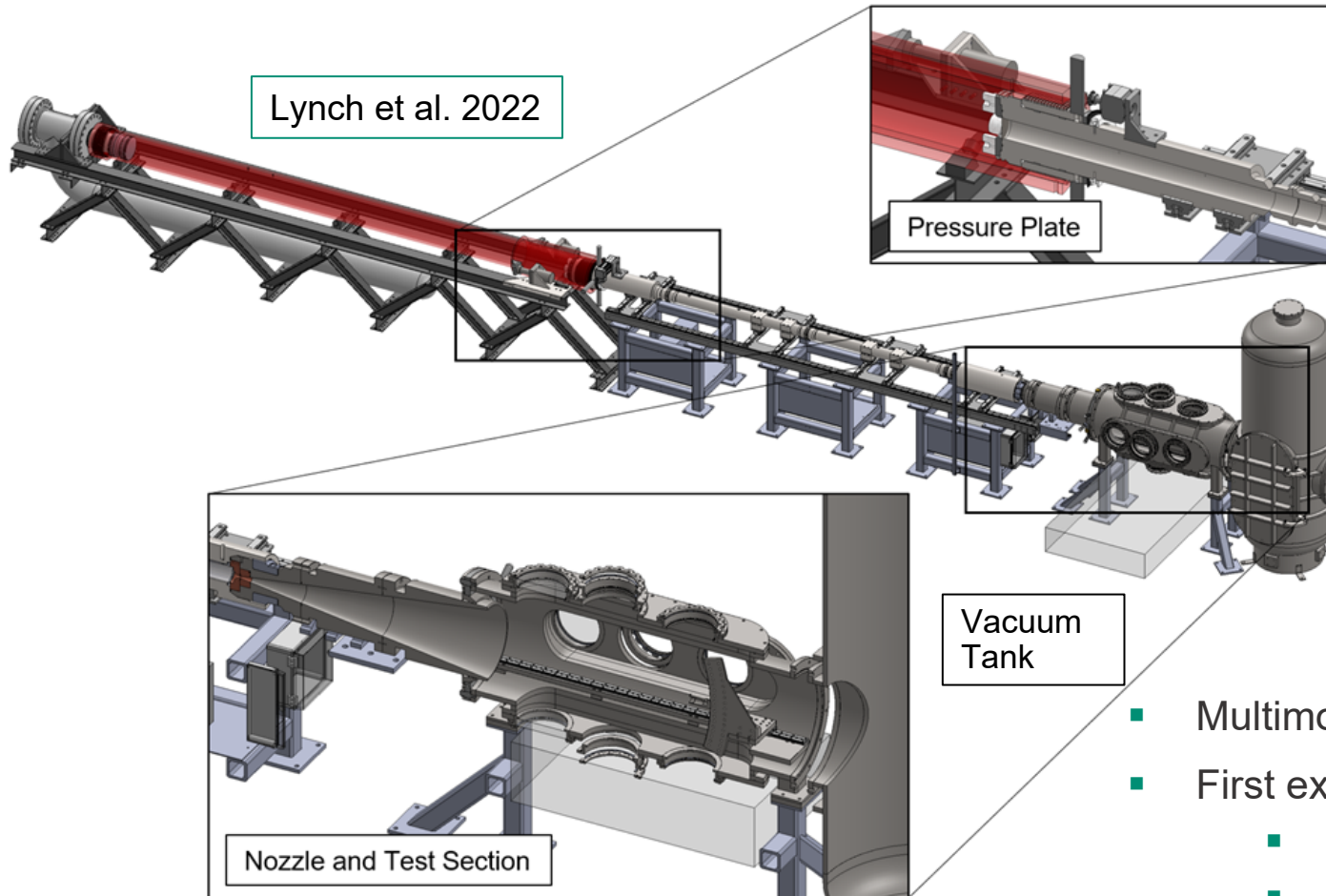


iPhone movie of  
NOPO spectrum  
tuning with 10-Hz  
pump laser

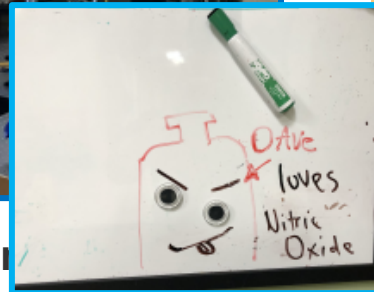
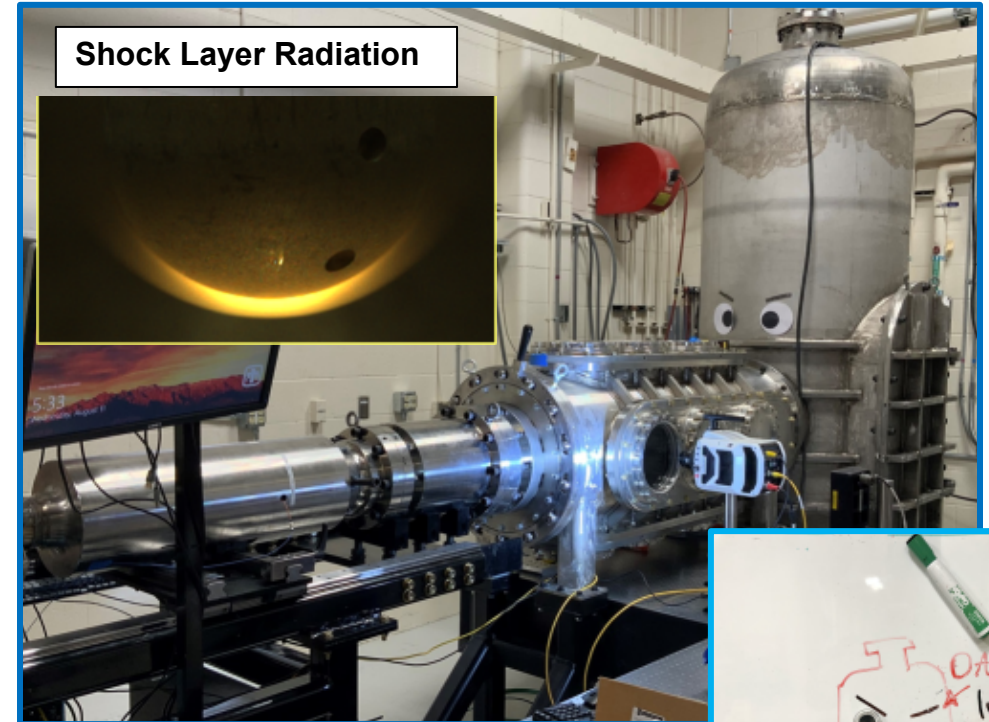
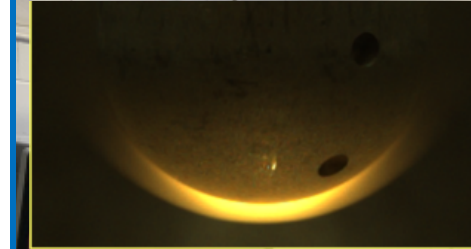
# Hypersonic Shock Tunnel (HST) Provides the Reentry Flight Environment



Lynch et al. 2022



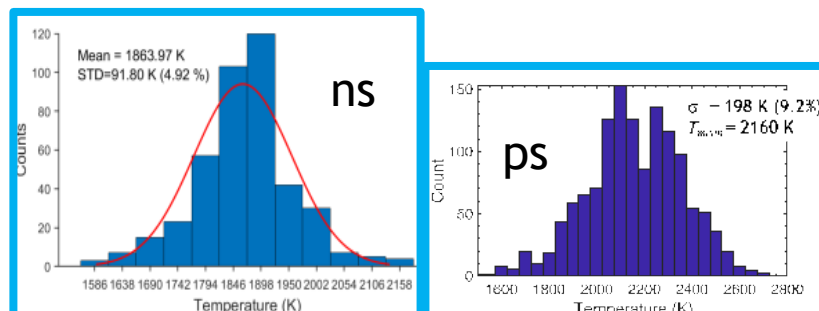
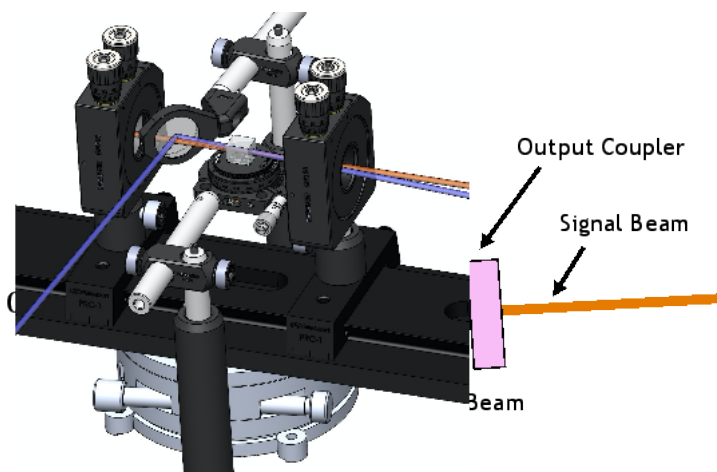
Shock Layer Radiation



- 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



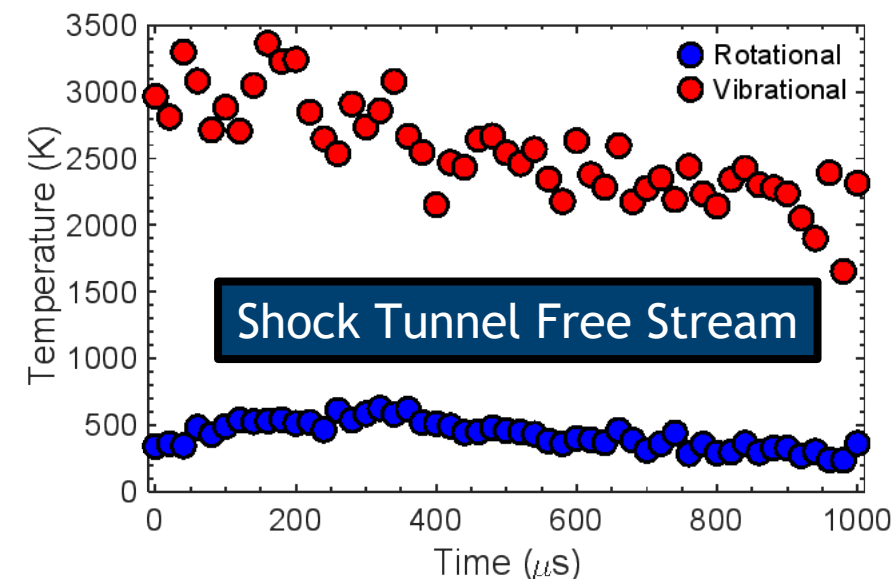
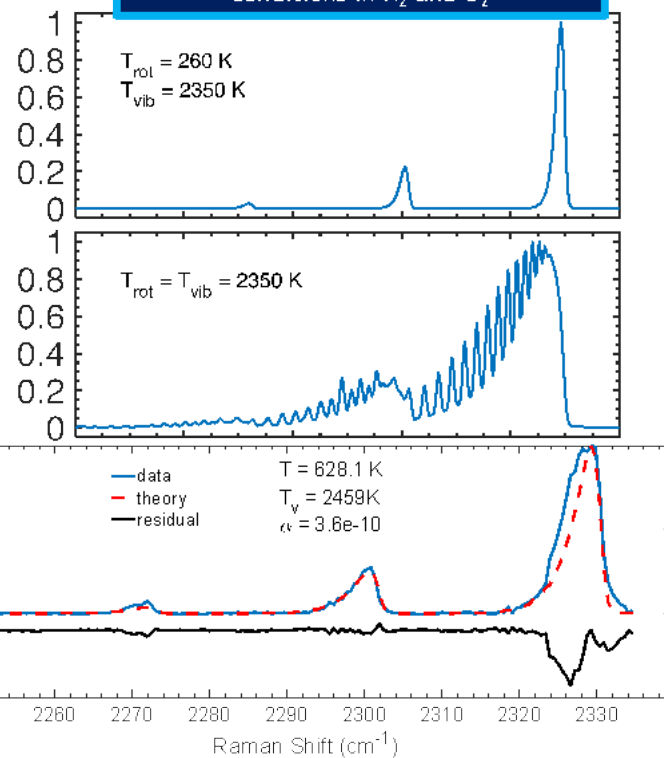
# Improved high-speed thermometry: 100-kHz *nanosecond* CARS



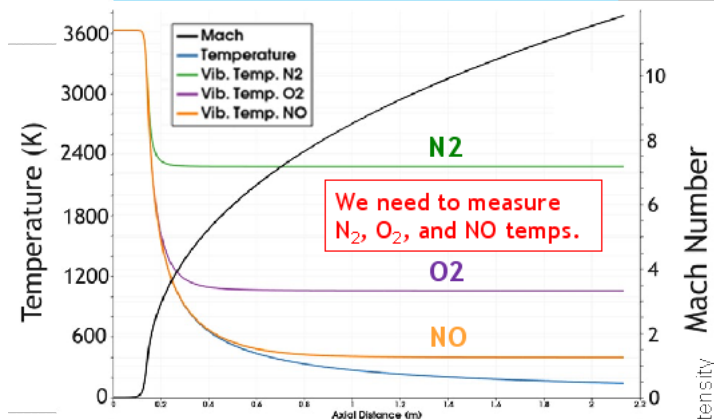
2X reduction in uncertainty!

- 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

CARS spectra can reveal *nonequilibrium* conditions in  $N_2$  and  $O_2$



SPARC Calculation of Nozzle Nonequilibrium



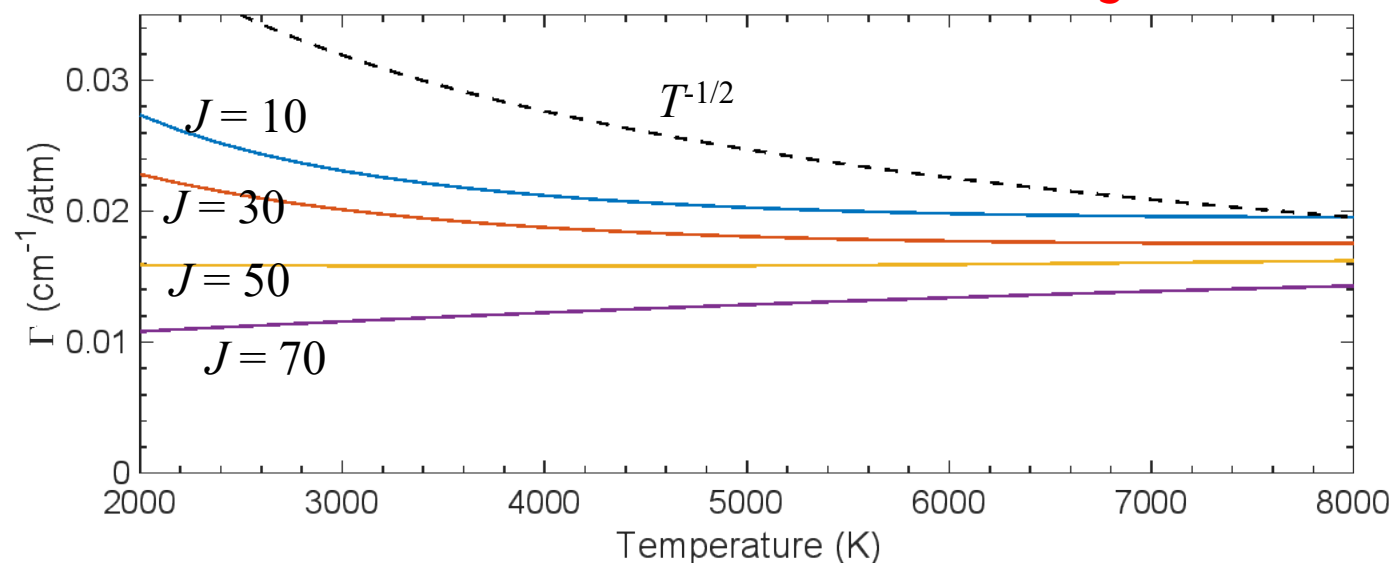
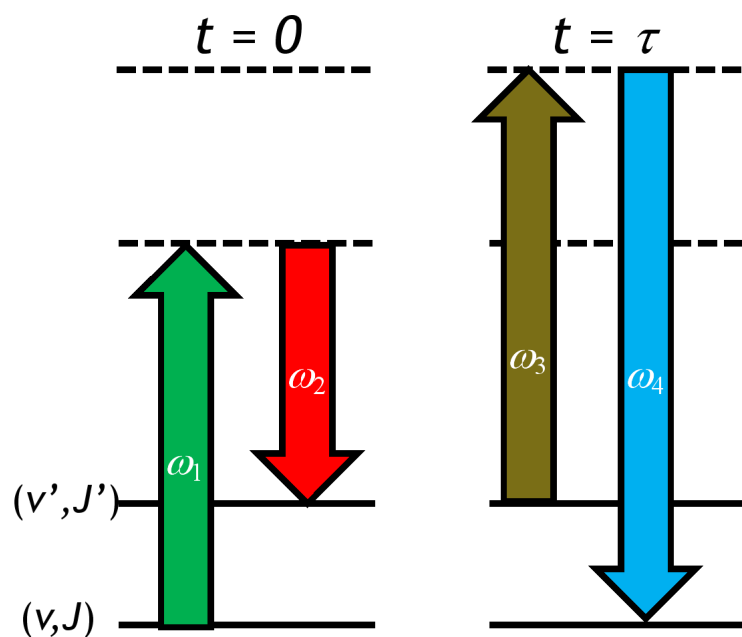
New SPARC models guide experiment design

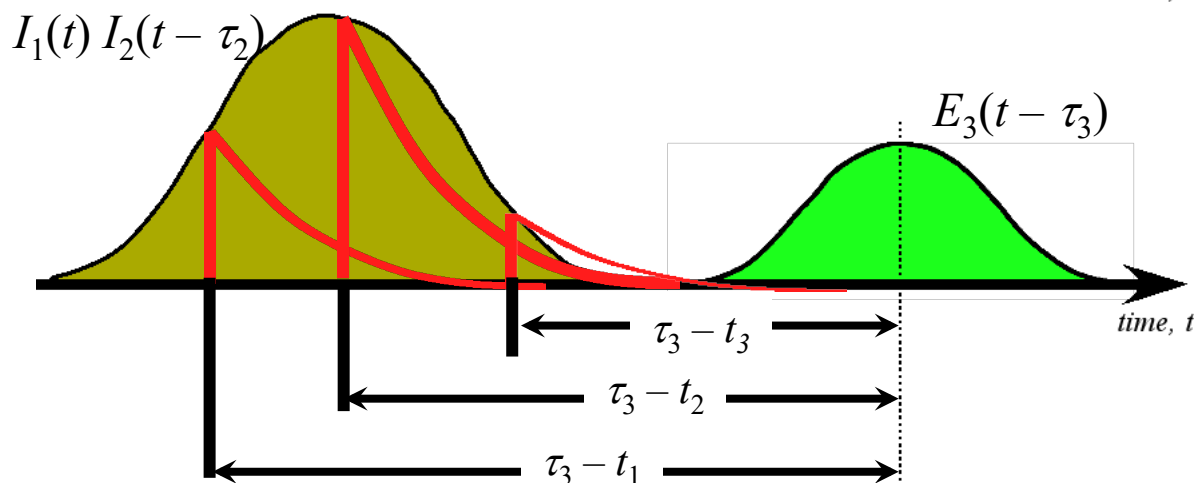
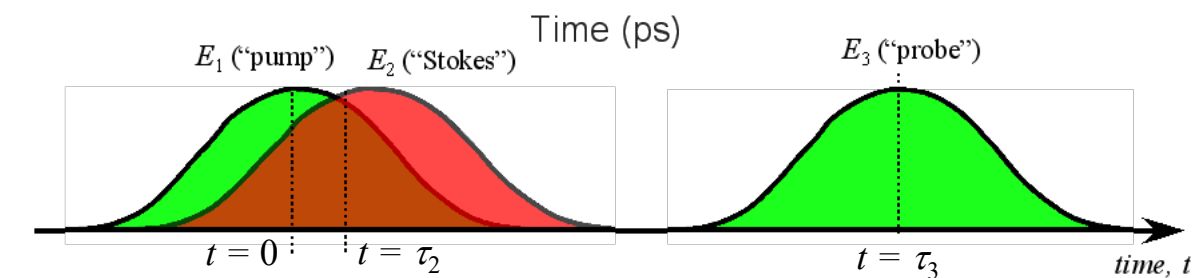
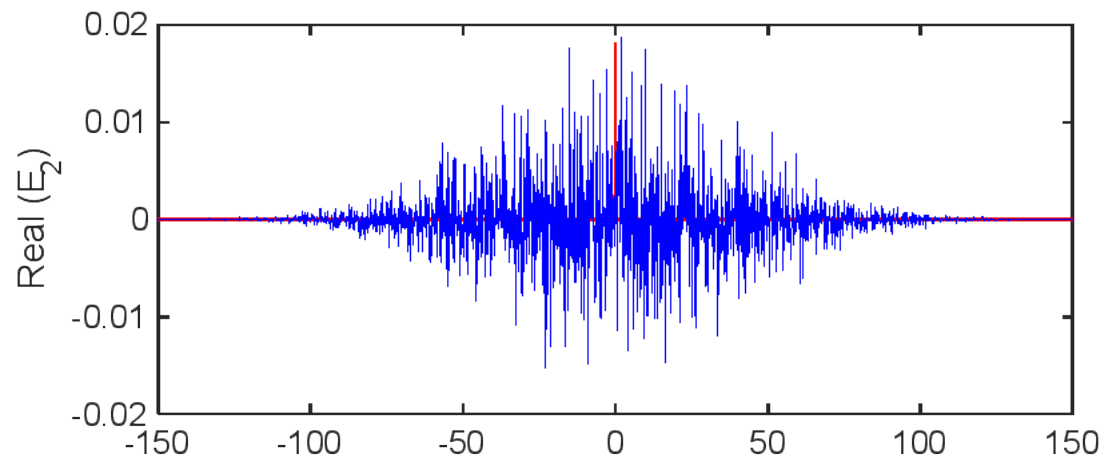
“Generalized Yuratich Equation,” Marrocco, *Opt. Lett.* 39, 4831 (2014)

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

Phase factor  
(time delay)

Extrapolate MEG  
linewidth model to very  
high T





$\ll 1$  fs (impulsive) CARS       $\sim 1$  ps CARS       $\gg 1$  ns (cw) CARS



### 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  $N_2$  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

