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Hybrid fs/ps CARS and Plasma Coupled-Combustion at Sandia

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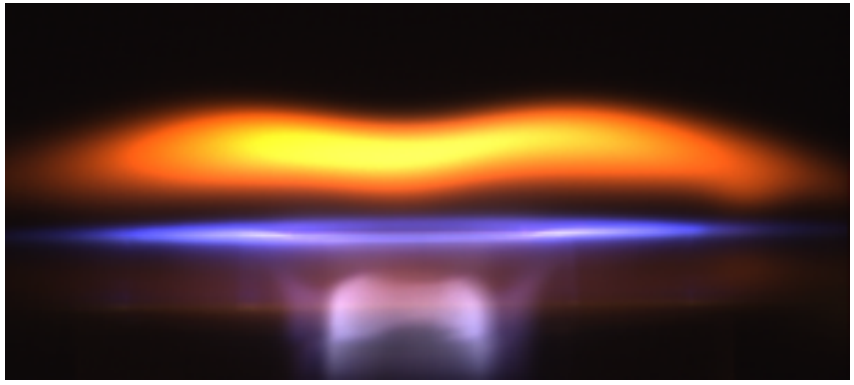

Sandia National Laboratories

PCI CSE Parallel Computing Institute Computational Science & Engineering

THE OHIO STATE UNIVERSITY

Center for Exascale Simulation of Plasma-Coupled Combustion XPACC

Hybrid fs/ps CARS

- CARS is a 4 wave mixing scheme
  - **Pump Beam**: Chosen to provide necessary frequency difference with stokes beam
  - **Stokes Beam**: Provides the bandwidth for the measurement
  - **Probe Beam**: Frequency narrow, used to scatter off the coherence
  - **CARS Signal**: Blue shifted off the probe beam
- What are you actually doing to the molecule?
  - Pump and stokes beams interact with molecules whose dipoles are aligned with the polarization vector of the beam, creating an initial alignment of the species
  - The molecules continue to rotate, with the period of realignment inversely proportional to the rotational constant of the species

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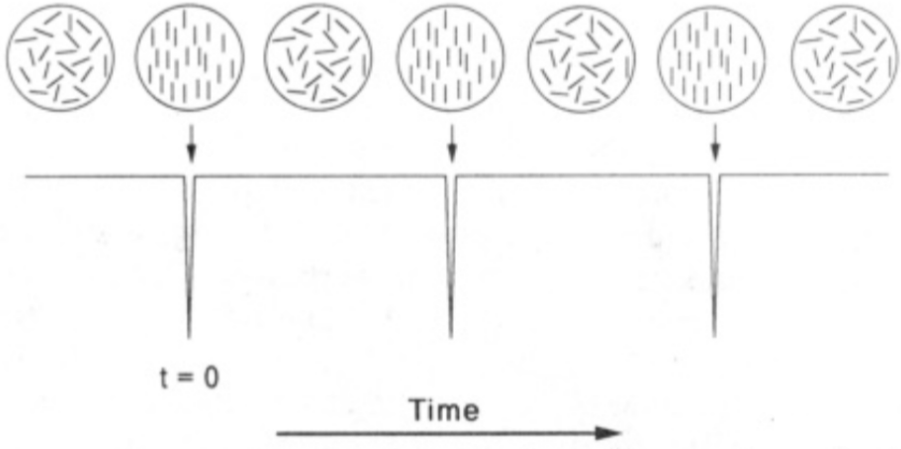
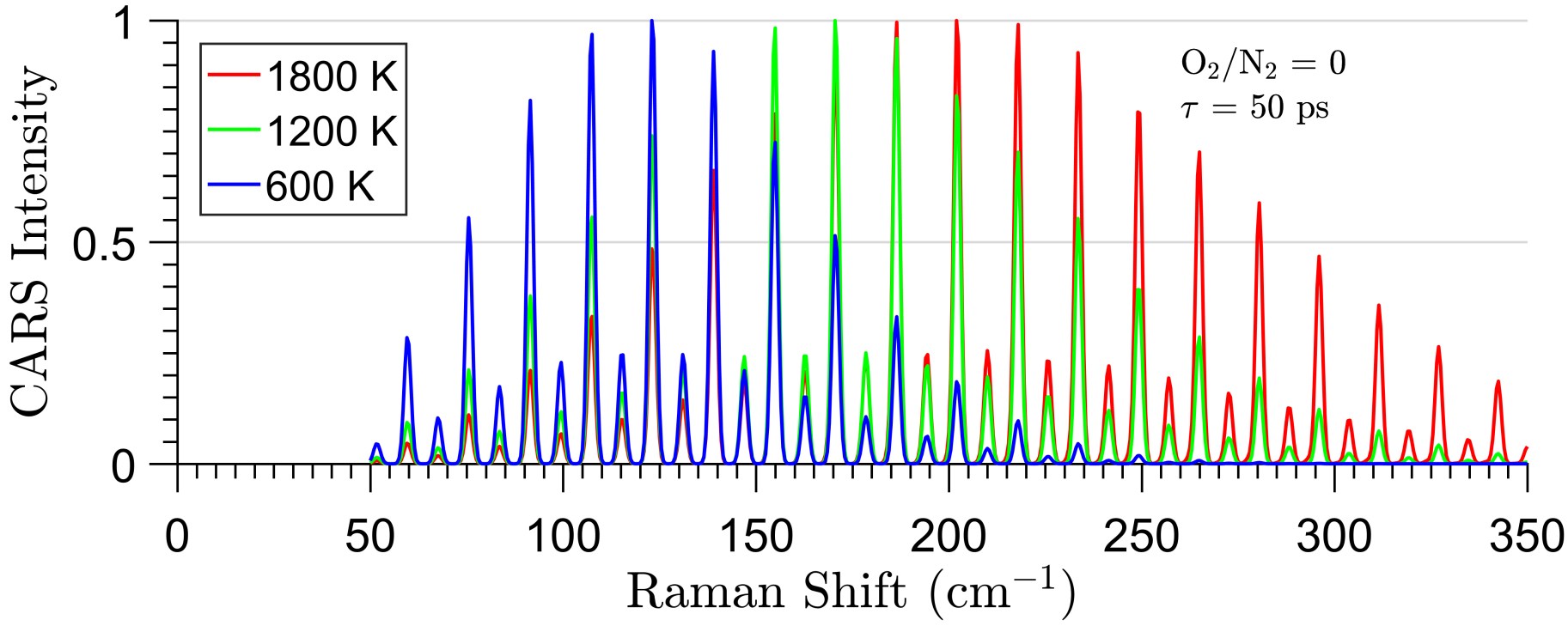
  - Felker, Journal of Physical Chemistry, 1992



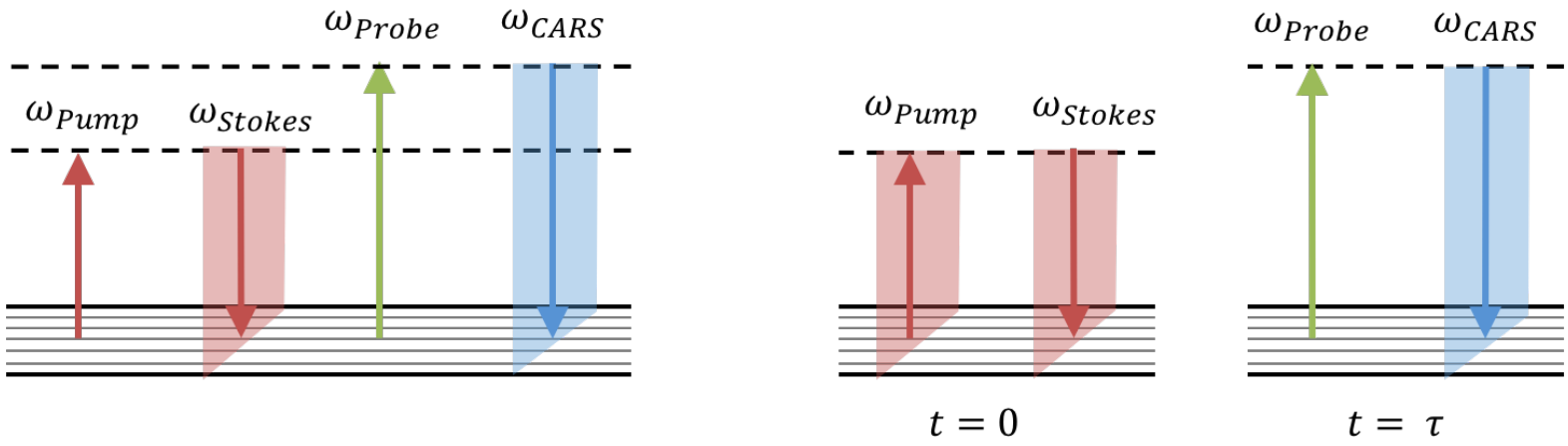
N_2 Rotational CARS Spectrum

- Higher temperature, populating more/higher rotational energy levels



Delaying the Probe Pulse

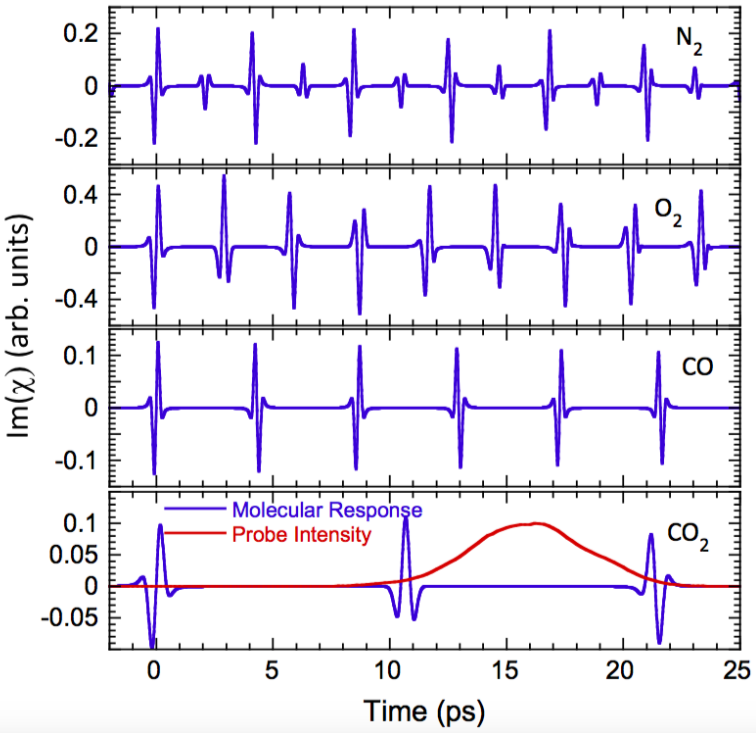
- Prepare Raman coherence in the molecule(s) of interest
  - Impulsively via femtosecond pump and stokes beams
- Probe coherence at some Δt from the pump/stokes beam
  - Frequency narrow probe beam



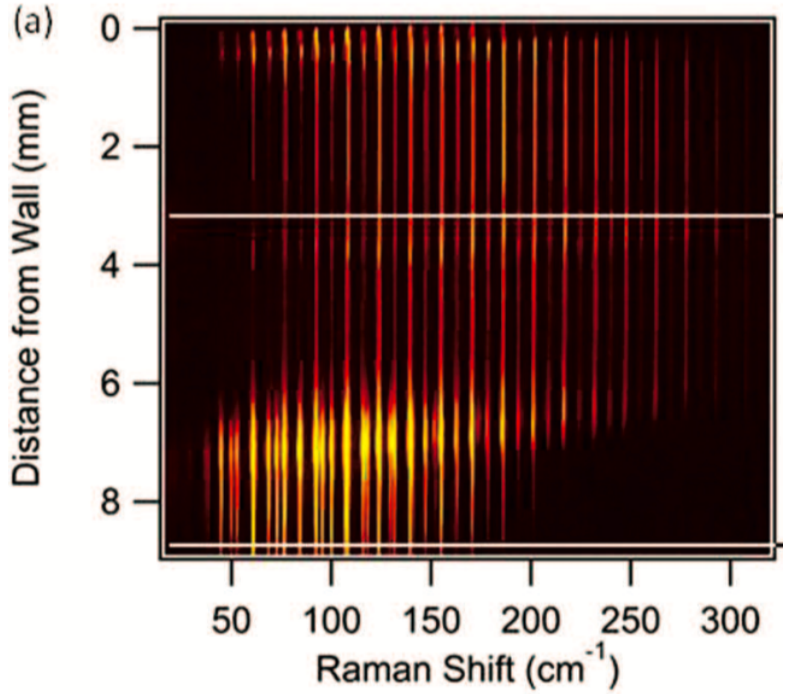
Delaying the Probe Pulse

- Benefits
  - Eliminate non-resonant background
  - Temporally select species of interest
  - Dynamic range
  - Collisional modeling (con?)

Kearney Combustion and Flame 2015



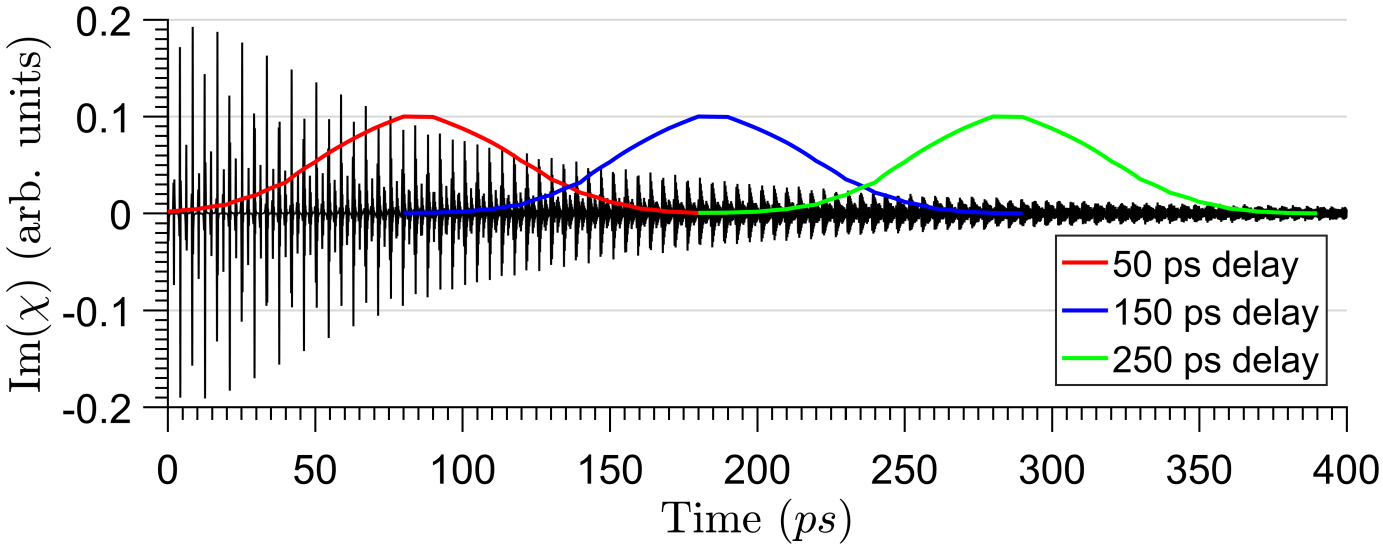
Bohlin et al. The Journal of Chemical Physics 2013



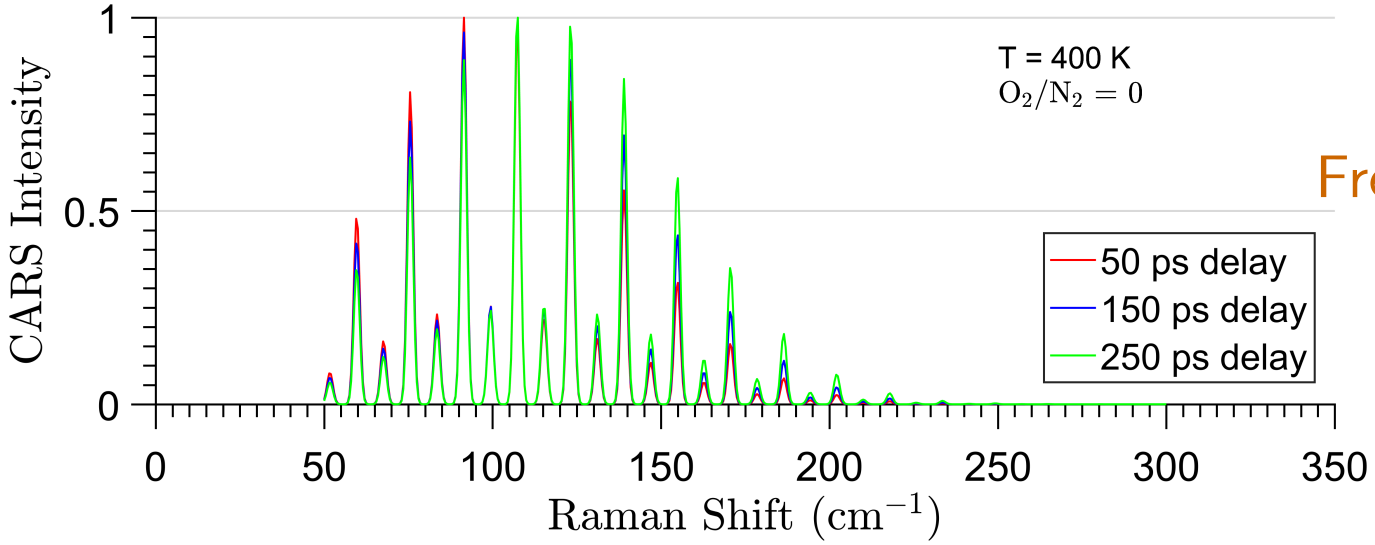
Fast CARSFIT Code

- Model molecular response in the time domain for a given probe pulse and delay
  - Boltzmann fraction (i.e. temperature)
  - Molecular constants
  - Collisional de-phasing
- Take fft of resulting signal, convert to wavenumbers
- Generate a 3D library of spectra
  - Temperature
  - O_2/N_2 ratio
  - Slit width or H_2/N_2
- Fit experimental spectrum to the library (4th order lagrange interpolation)
  - Residual norm cutoff

Delaying the Probe Pulse



Time Domain



Frequency Domain

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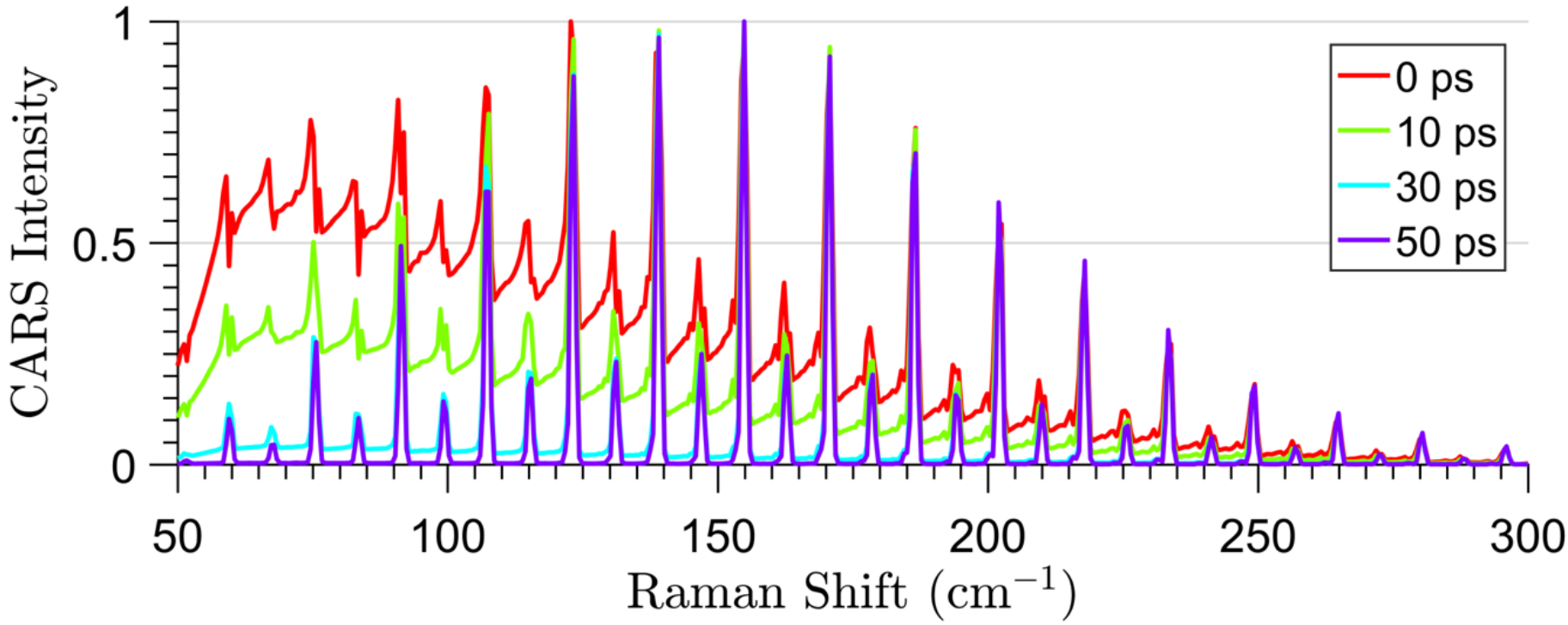
Delaying the Probe Pulse

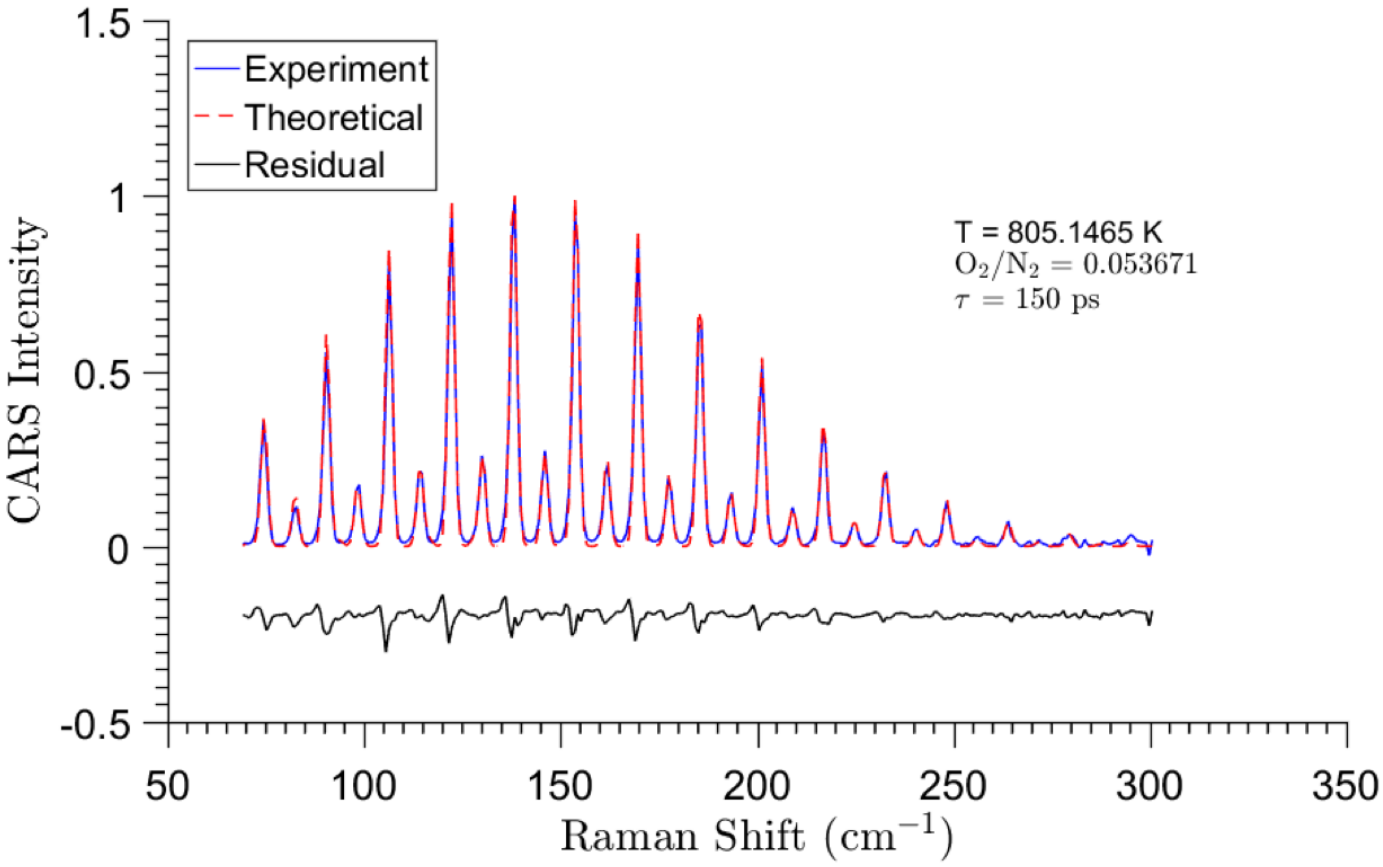


Experimental Processing

- Collect the following
  - Signal
  - Non-resonant spectrum (beams crossing in argon)
  - Background for both (block the stokes beam)
- (Signal - Background)/(Argon - Background)
- Pixel calibration against synthetic CARS spectrum
- Least squares fit the result against a precomputed library
- Hydrogen concentration
  - Ignore hydrogen lines, fit for temperature and O_2/N_2
  - Fix temperature and O_2/N_2, fit for hydrogen concentration

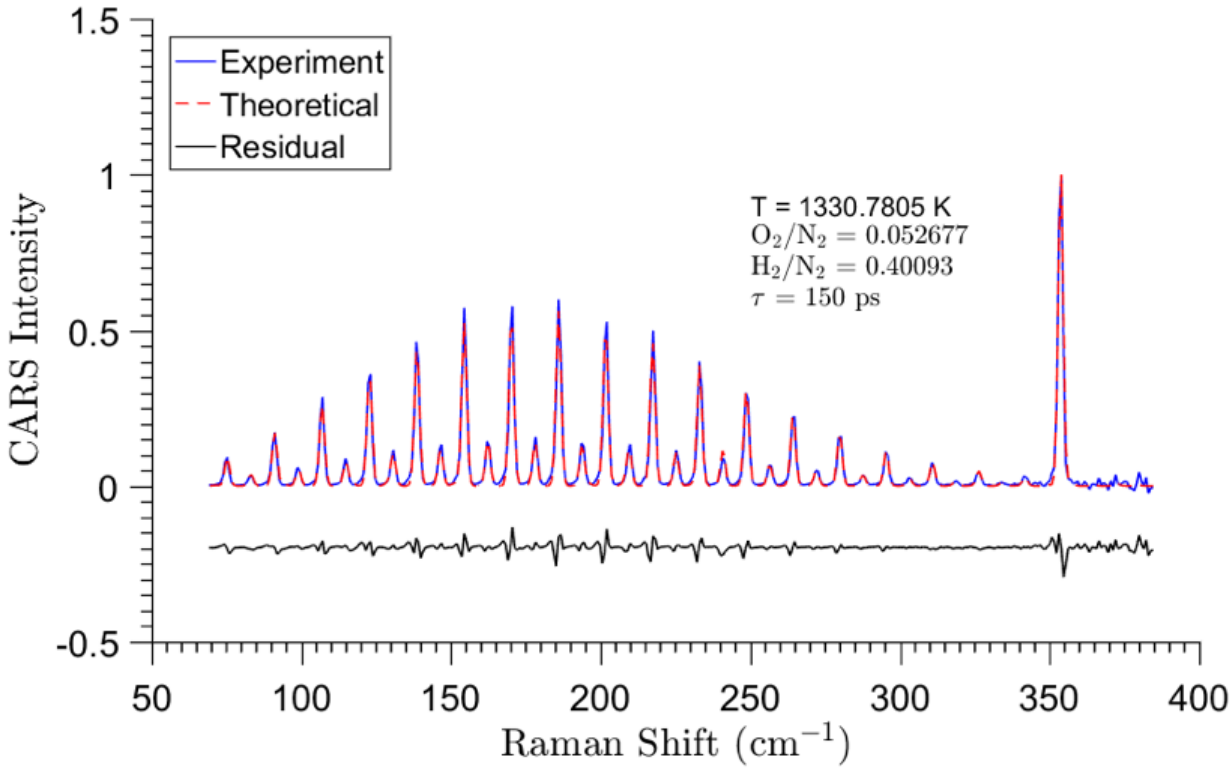
Examples of Fits

- Temperature
- O_2/N_2 Ratio

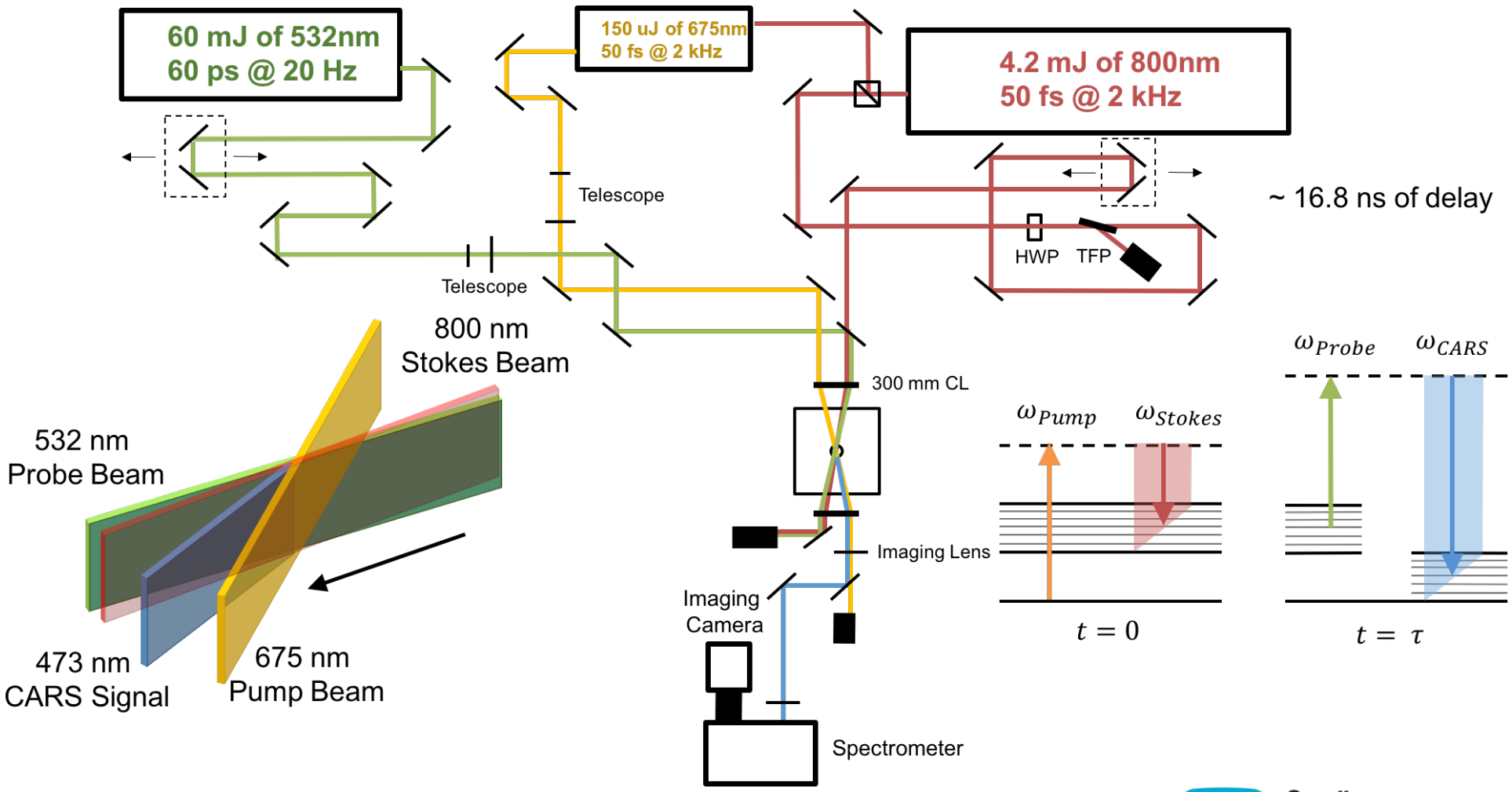


Examples of Fits

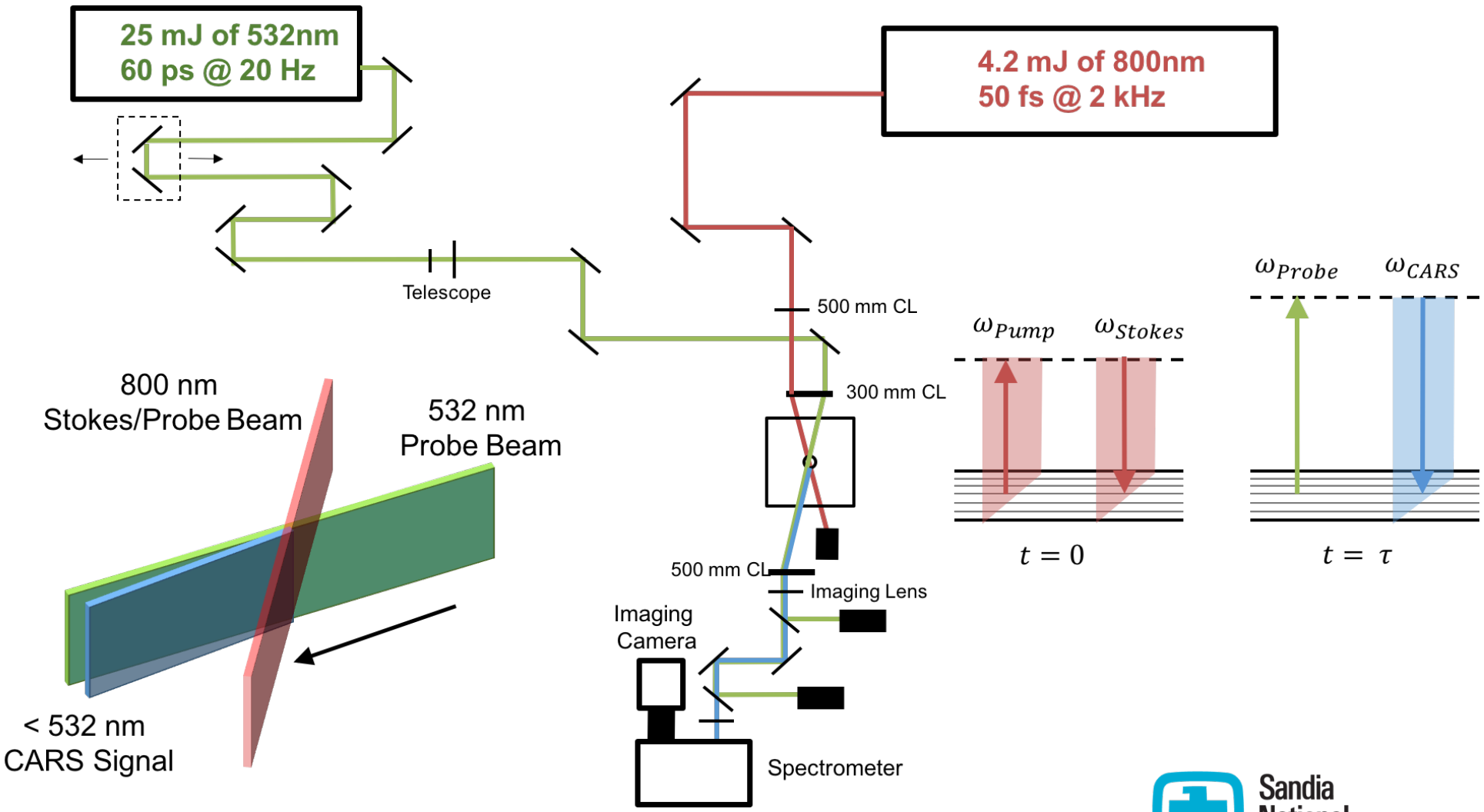
- Temperature
- O_2/N_2 Ratio
- H_2/N_2 Ratio



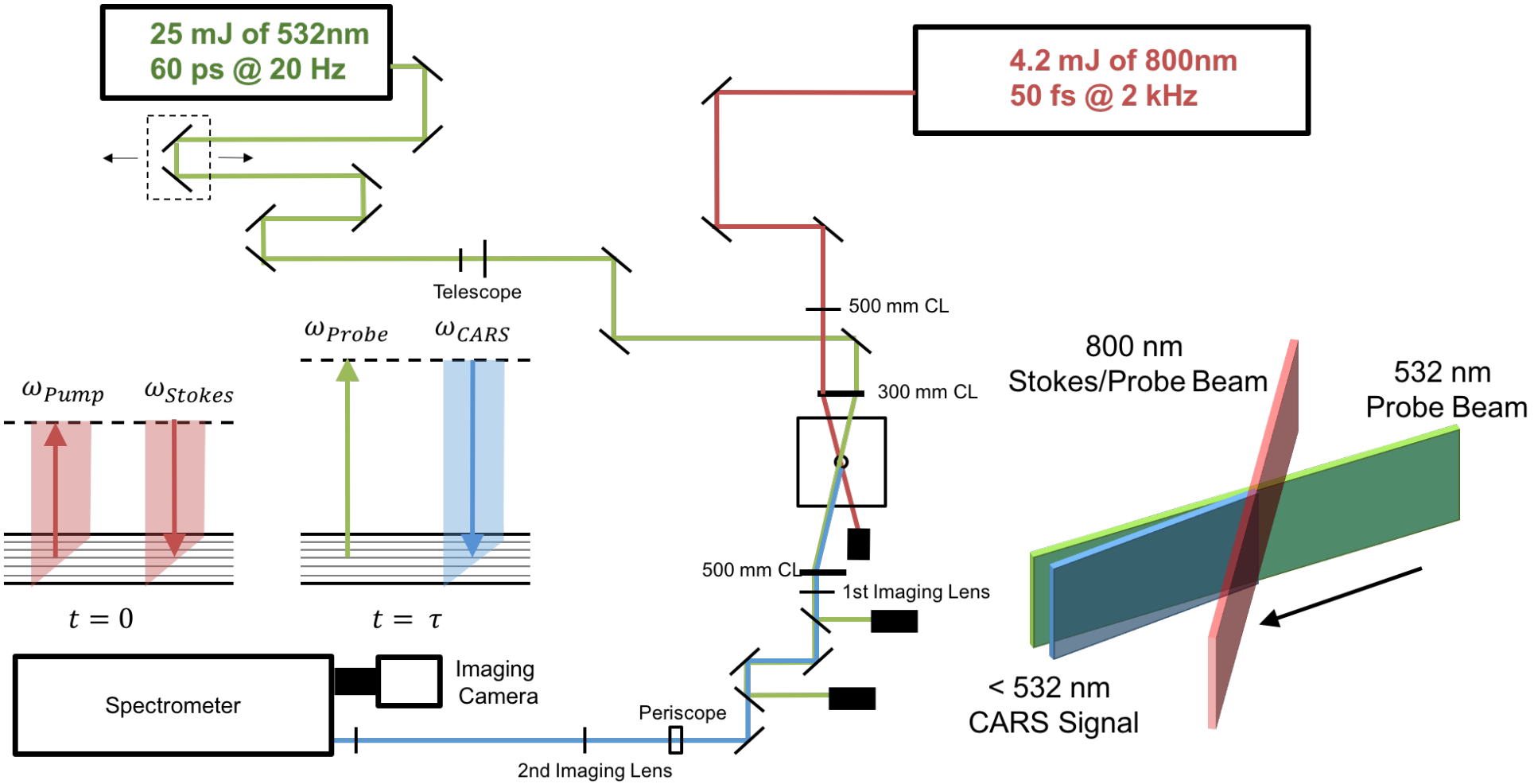
N_2 Vibrational 1D CARS



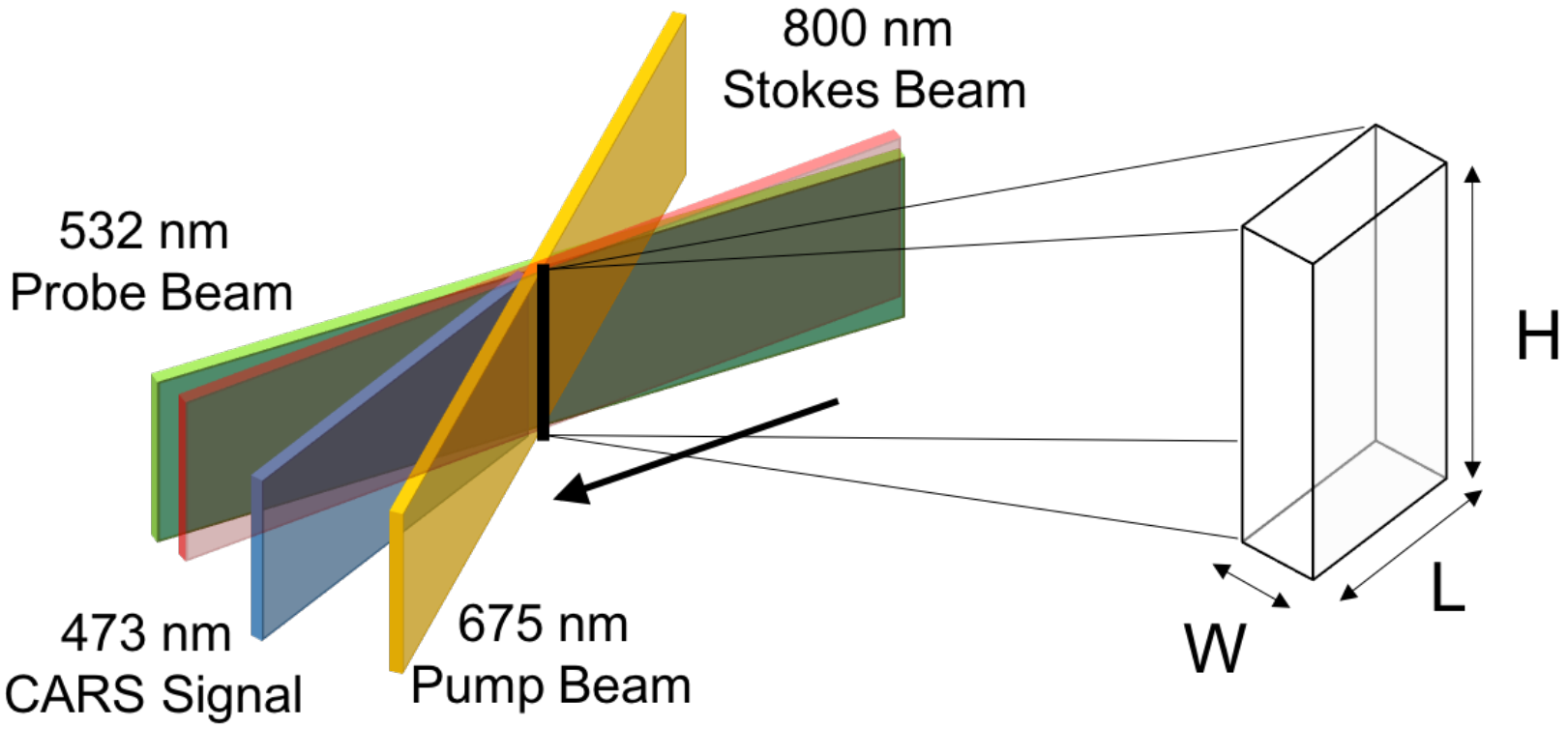
N_2, O_2, (and H_2?) Rotational 1D CARS



N_2, O_2, (and H_2?) Rotational 1D CARS v2.0

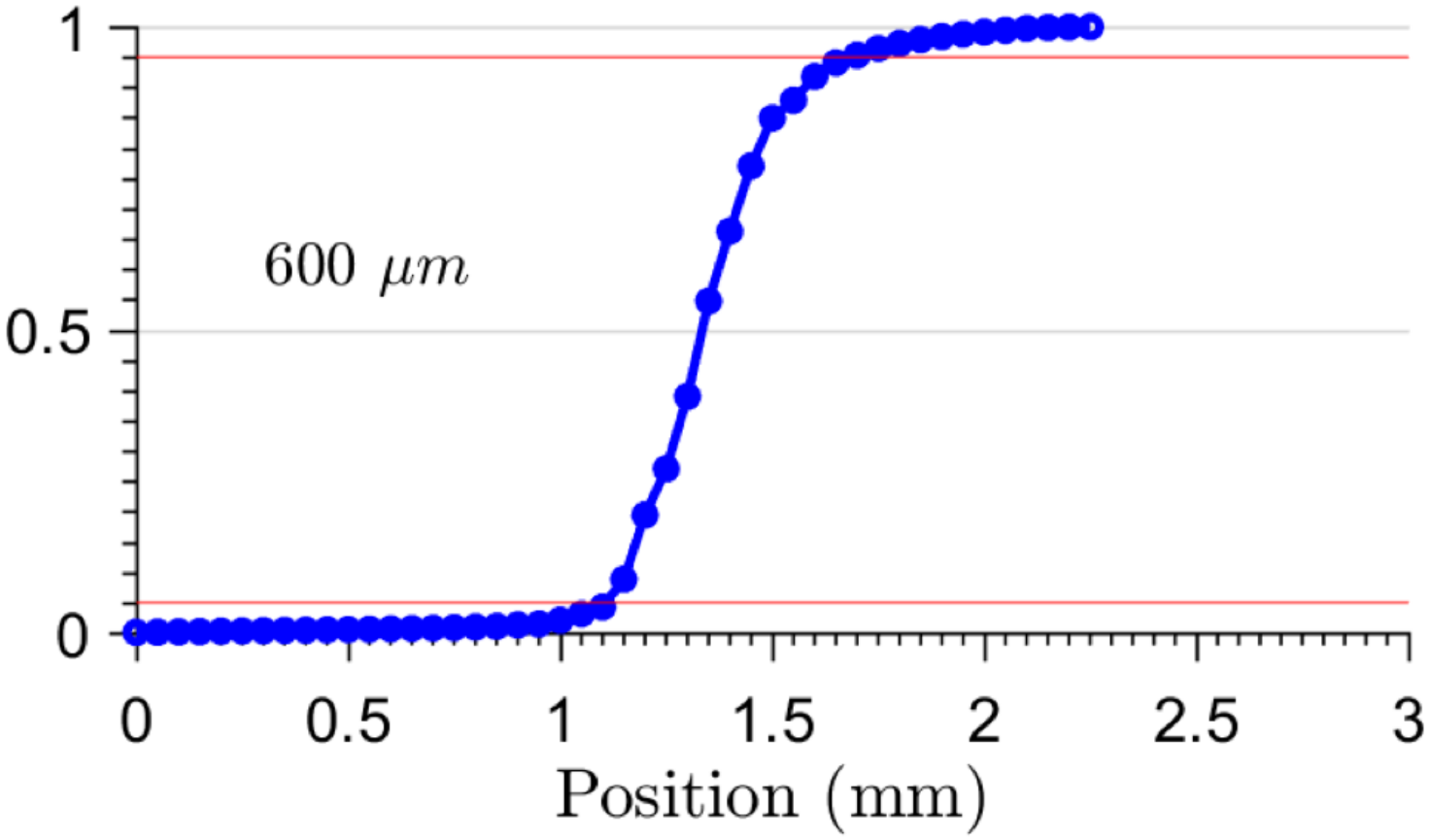


Measurement Volume



- H: Height of the shortest beam (pixels on detector)
- W: Beam waist at the focus (beam profiler)
- L: Length of signal generation (glass cover slip)

Measurement Volume



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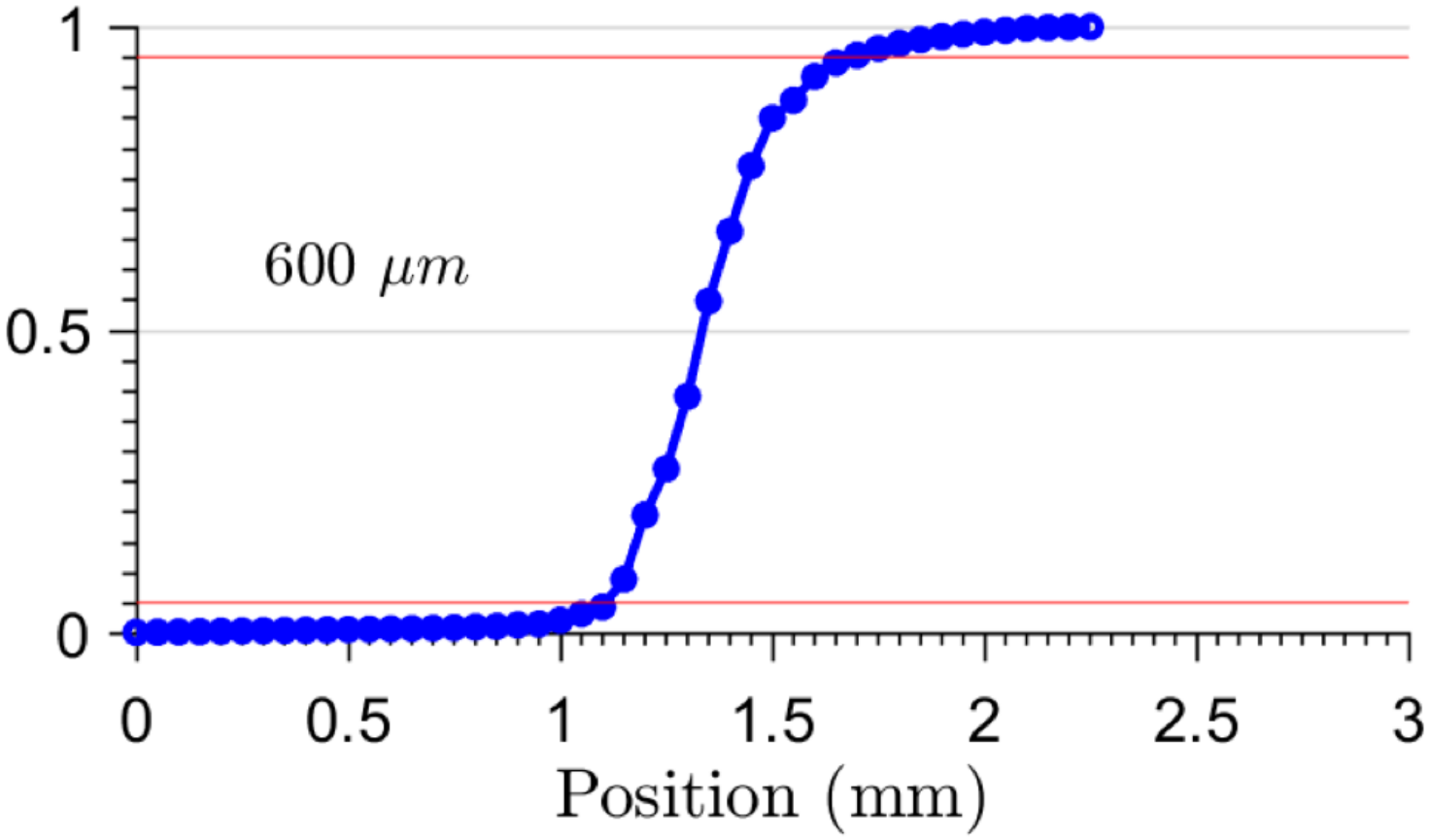
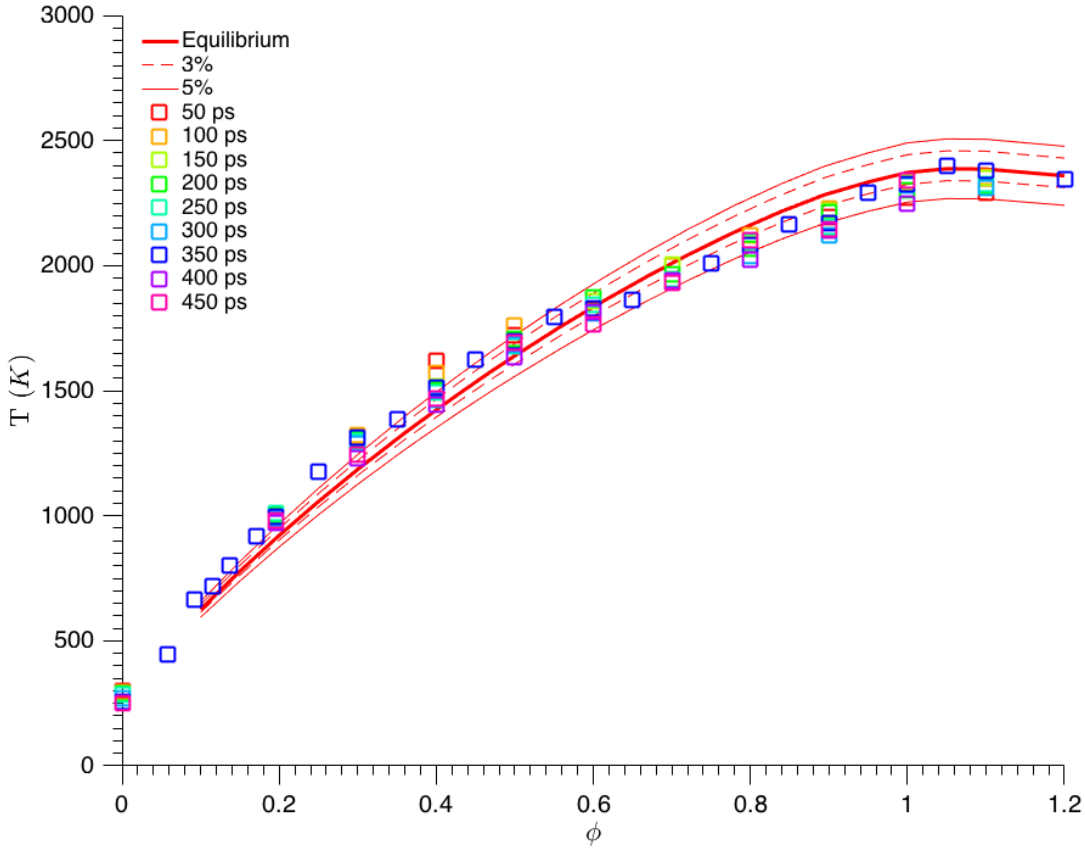


- H: 140 um
- W: 30 um
- L: 600 um

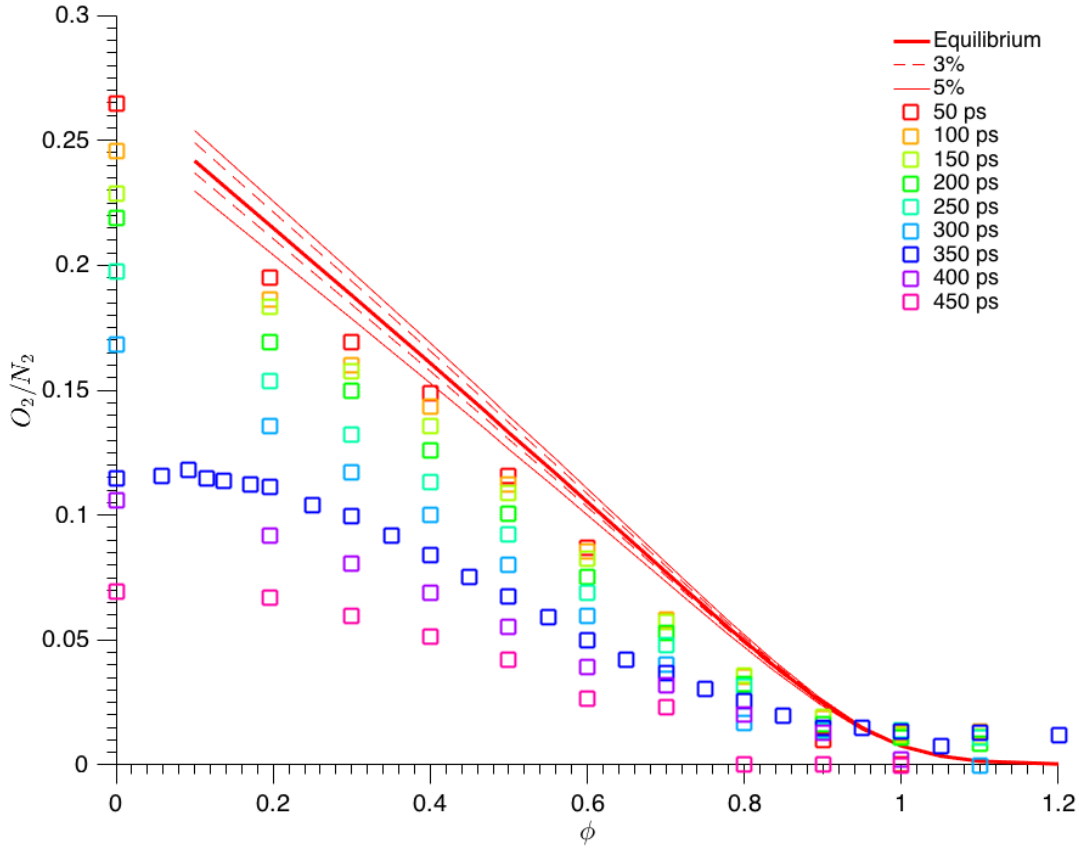
Hencken Burner

- Compare experimental results to equilibrium calculations
- These are average spectra results (for now...)



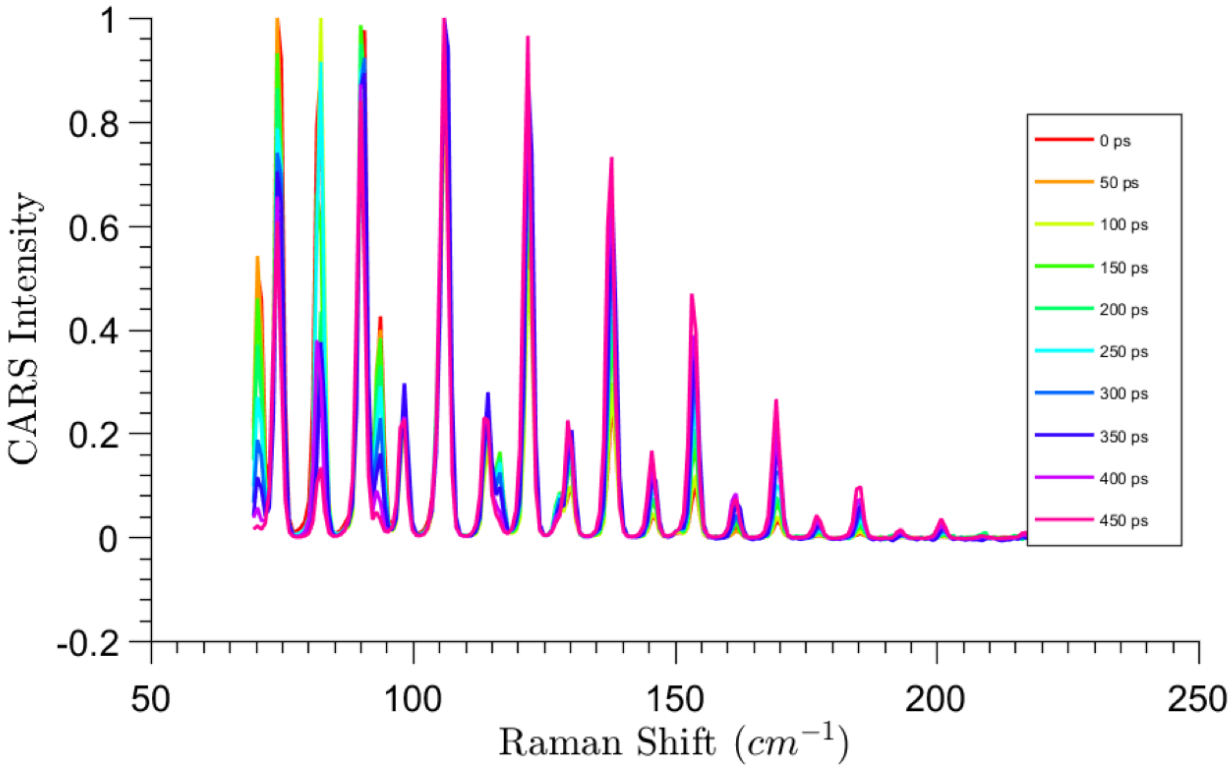
Hencken Burner

- Compare experimental results to equilibrium calculations
- Issues with O2 measurements...



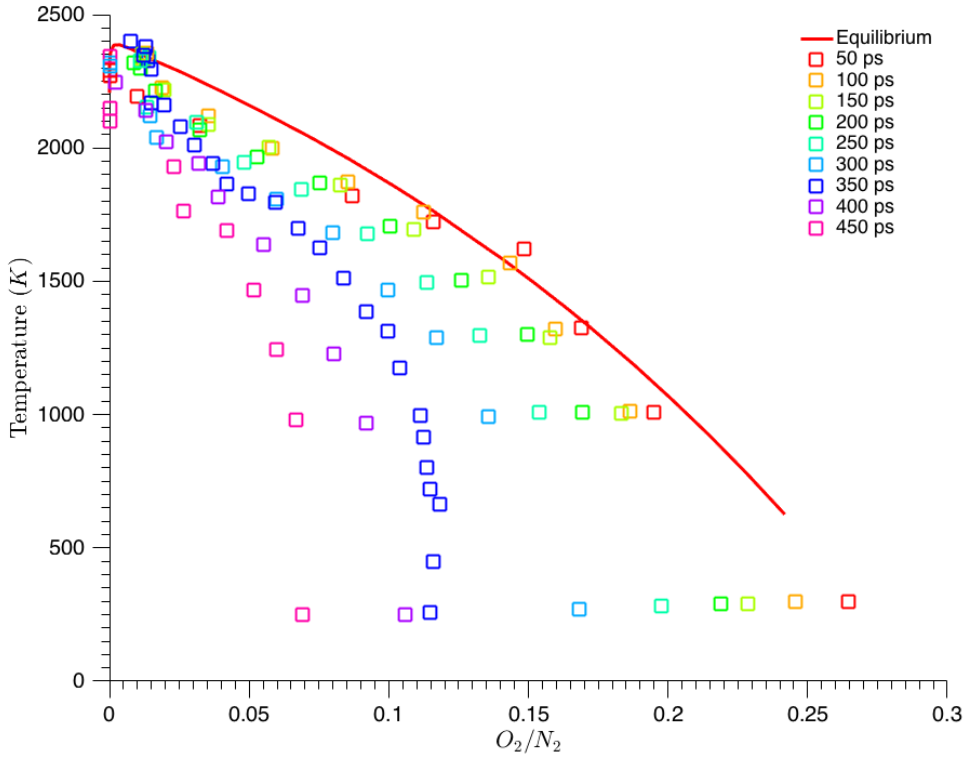
Room Air

- Measurements taken in air to see the oxygen content at longer probe delays



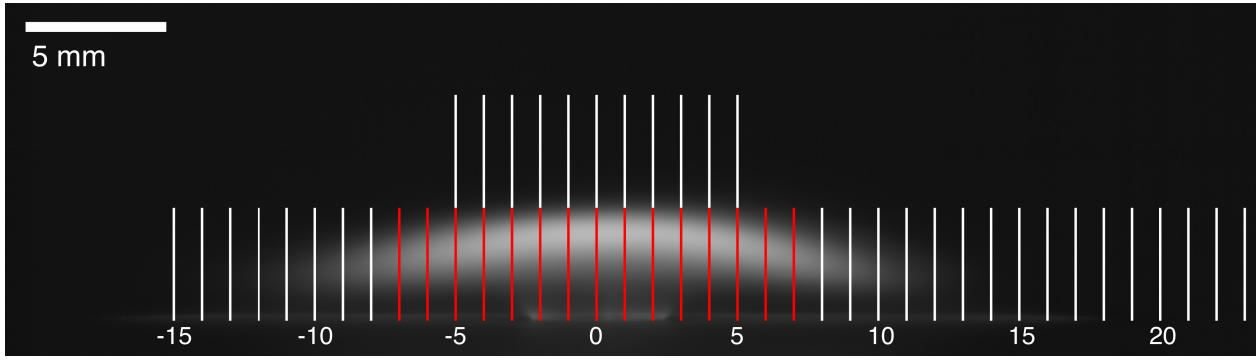
Collisional Model

- Oxygen is not modeled correctly at longer probe delays
  - O_2 linewidths?
  - Polarizability anisotropy?
  - Different for different probe delays? Can we defend this?



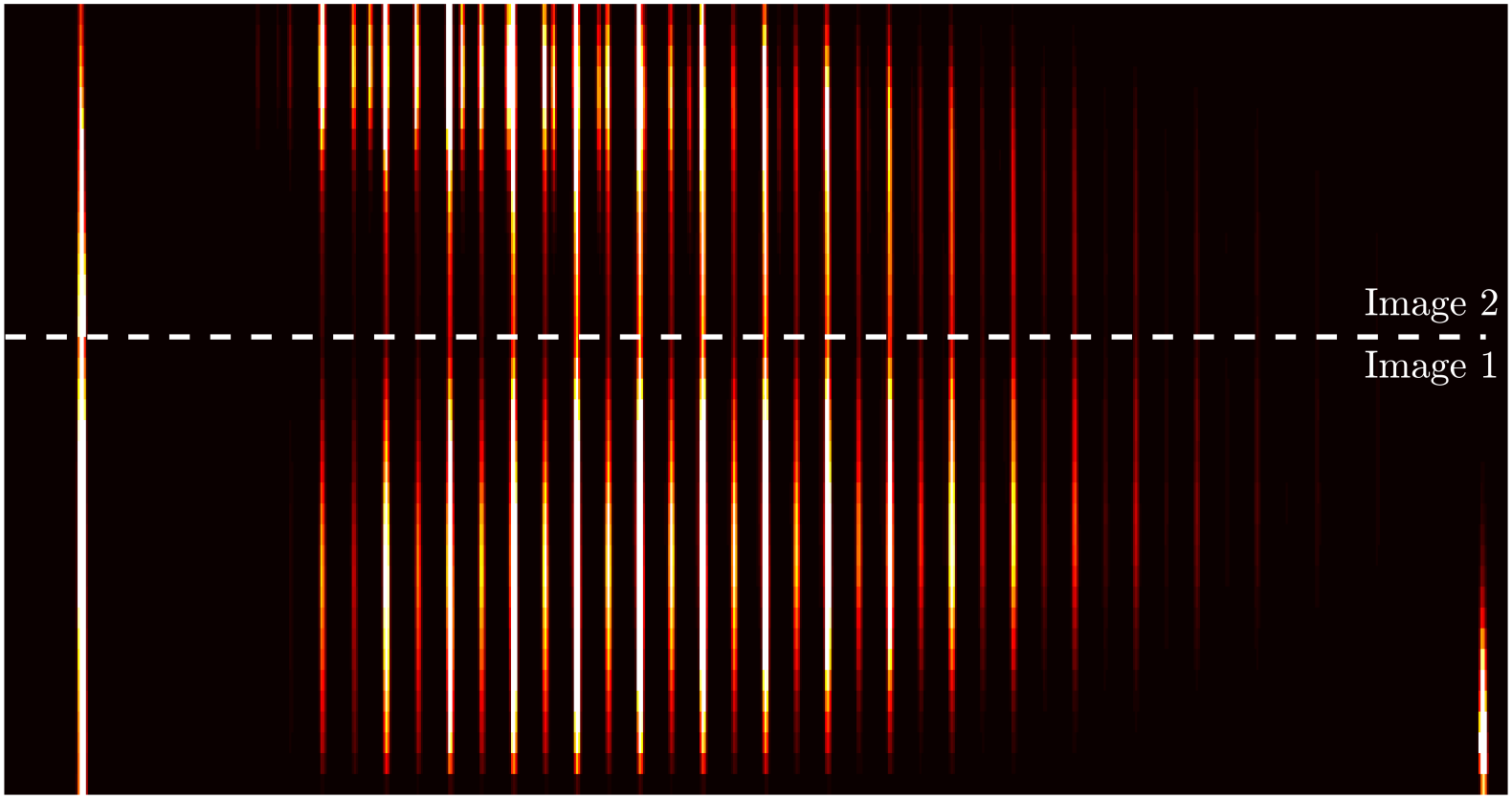
Experimental Conditions

- Measurement volume = 0.00252 cubic mm
- Acquisition at 20 Hz, vertically binning by 10, horizontal binning by 2
- Laser energies
  - 532nm: 24.8 mJ, 60 ps, 20 Hz
  - 800nm: 4.3 mJ, 50 fs, 2 kHz
- DBD Settings
  - 18 kHz
  - 8.75 kV peak



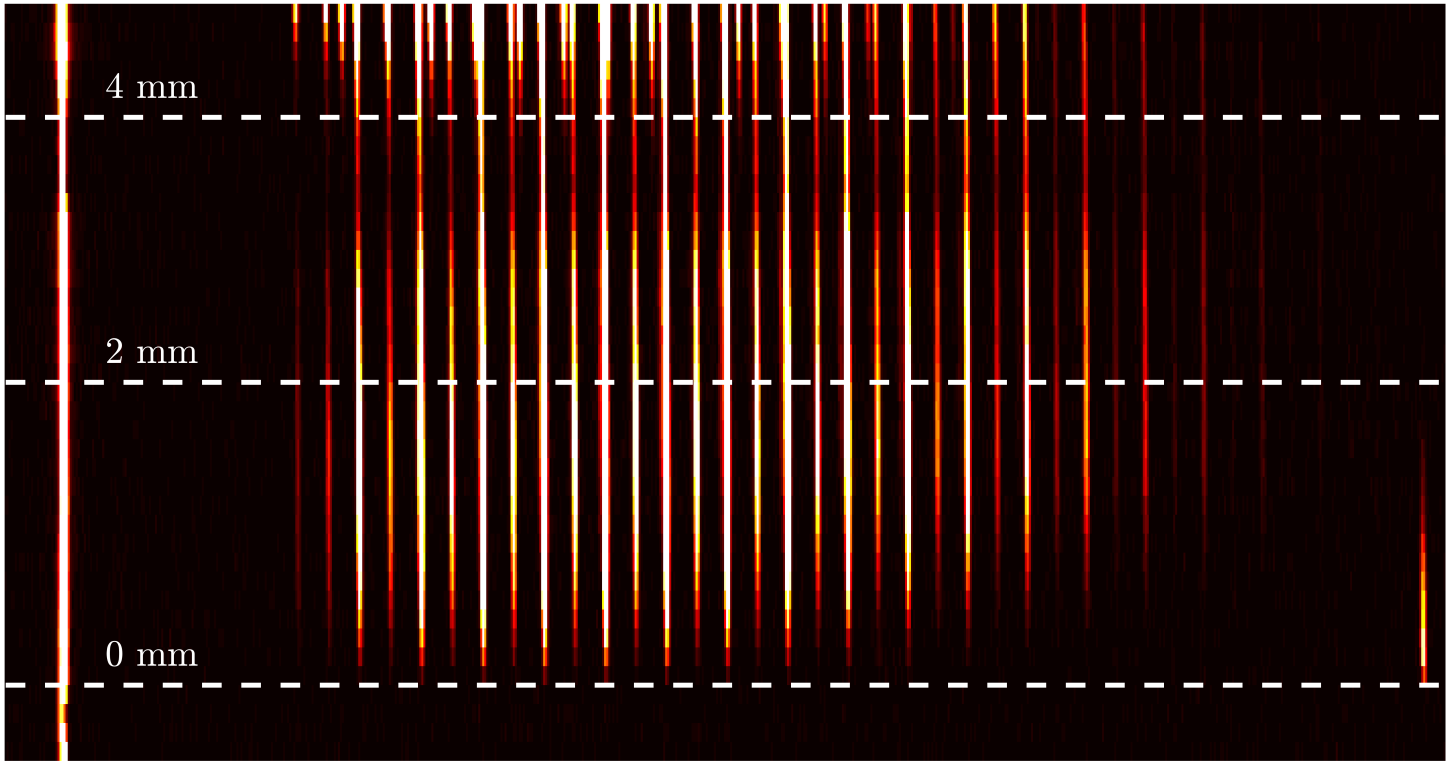
DBD Burner?

- Applied to the DBD Burner



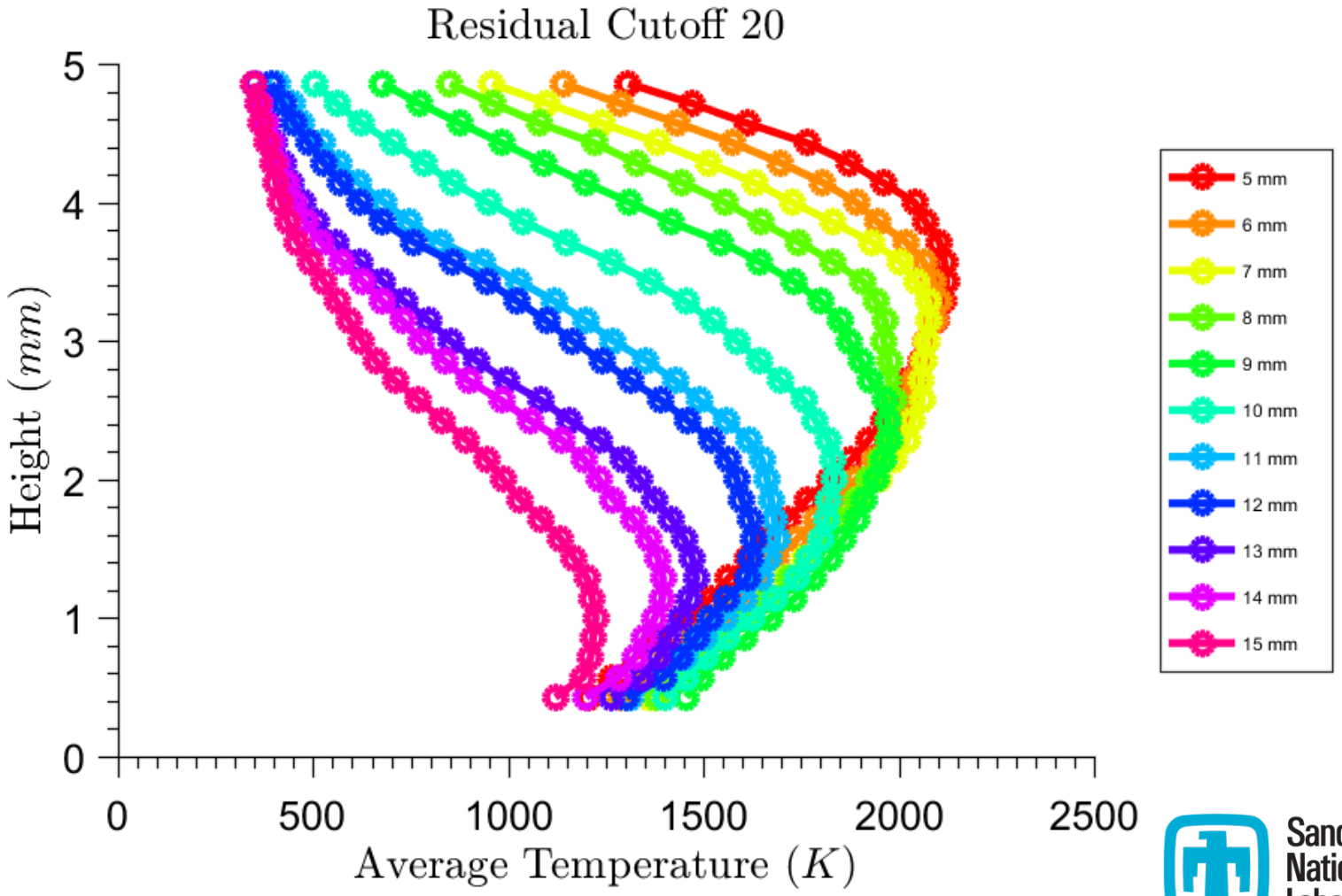
Single Shot Example

- Yes, this is one laser shot
- At the centerline (r = 0 mm)



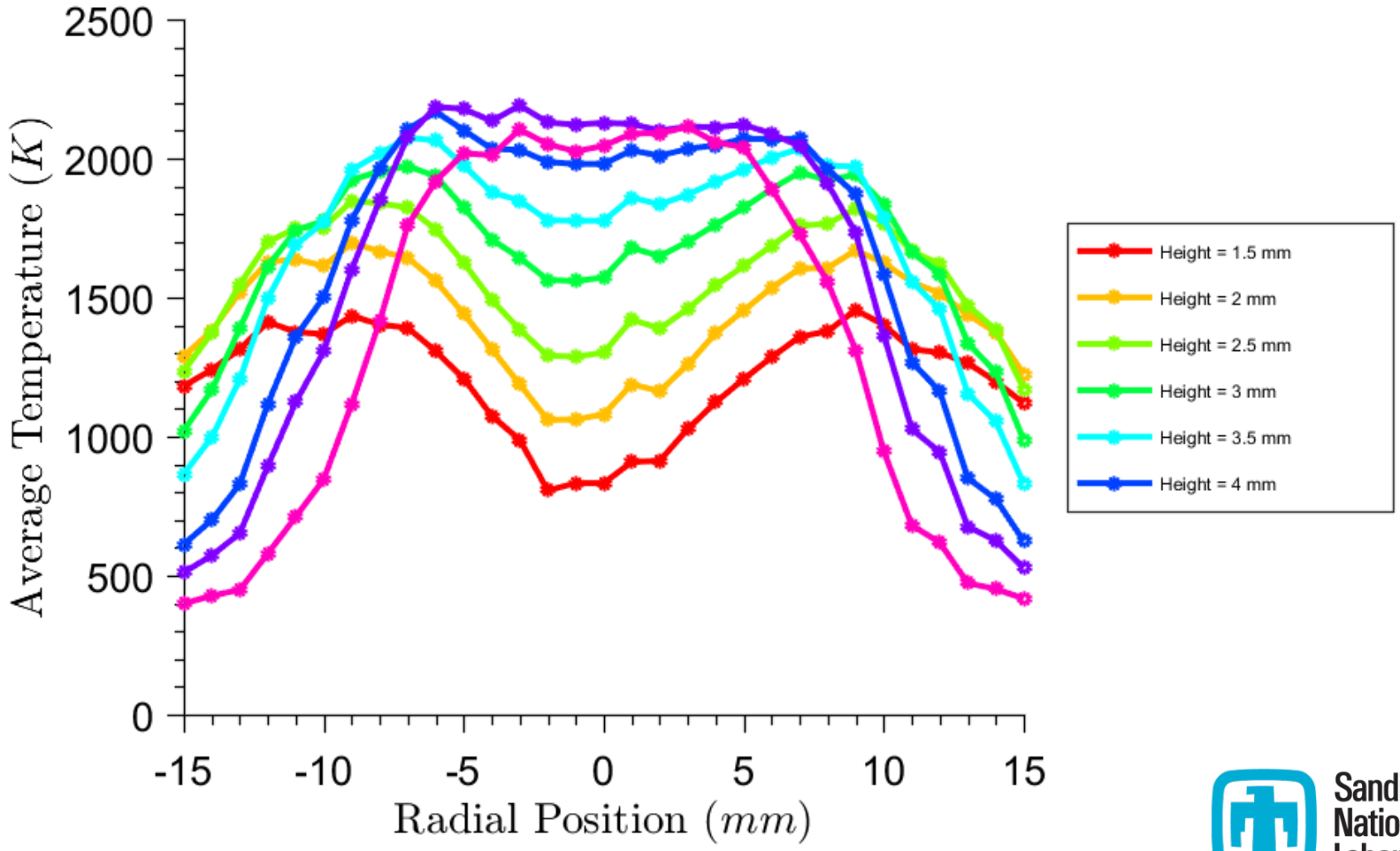
Preliminary Results

- Height v. Temperature for different radial locations



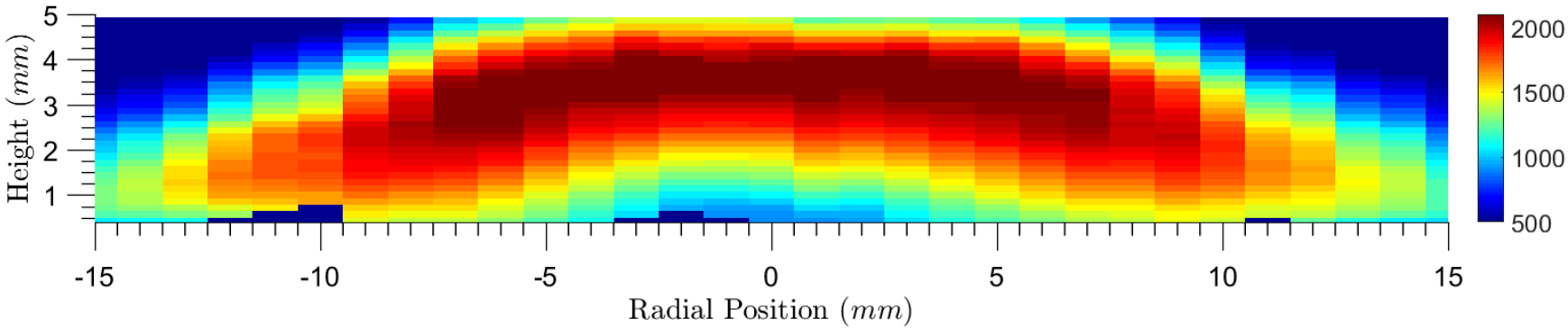
Preliminary Results

- Temperature v. radial position for different heights



Preliminary Results

- Contour plot of temperature



Acknowledgements

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