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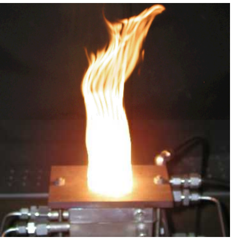

Hybrid fs/ps CARS for Sooting and Particle-laden Flames

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Overview

- Background
  - Sandia applications in “hostile” environments
- Ultrafast (fs/ps) CARS development
  - Why ultrafast CARS?
  - Hybrid rotational CARS
  - Hybrid vibrational CARS
- Measurements in metalized propellant burns with hybrid rotational CARS system
- Canonical Flame Measurements with vibrational CARS system
- Future work

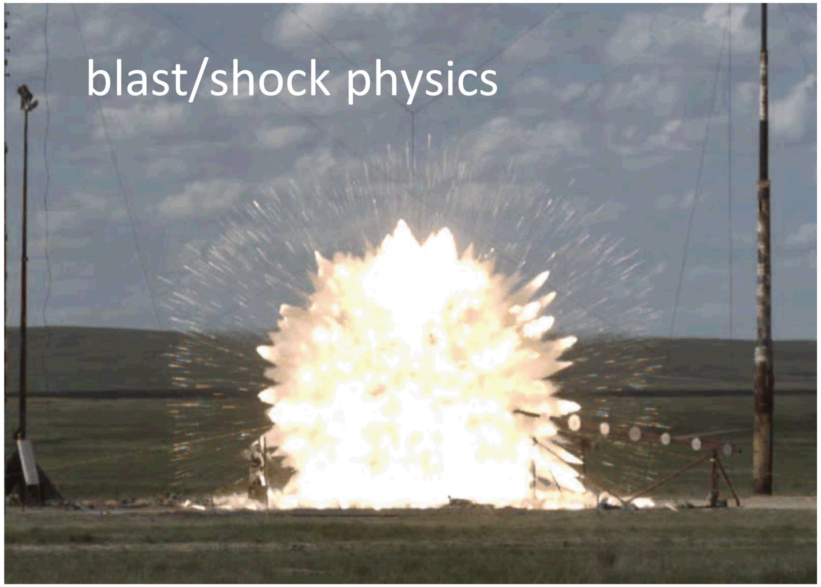
Contributions from . . .

- Sean Kearney
  - Nanosecond CARS measurements
  - Hybrid rotational CARS measurements
- Dan Guildenbecher
  - Digital holography
  - Propellants
- Lee Stauffacher
  - Propellant Burns
- Kate Hoffmeister (me)
  - Hybrid vibrational CARS measurements

Sandia National Security Mission

- DOE strategic systems safety
- DOD/WFO applications
- Challenging environments
- Large-scale
- Heat, blasts, particulates

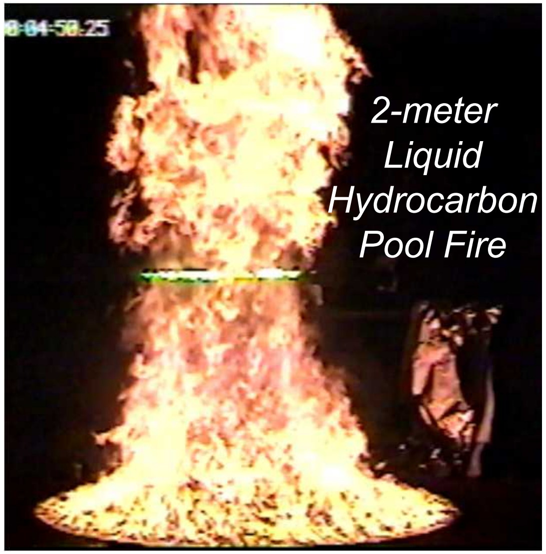
http://www.cbsnews.com/news/rocket-crash-no-immediate-threat-to-station-but-cause-is-unknown/



http://abcnews.go.com/Technology/Travel/wireStory?id=3529012

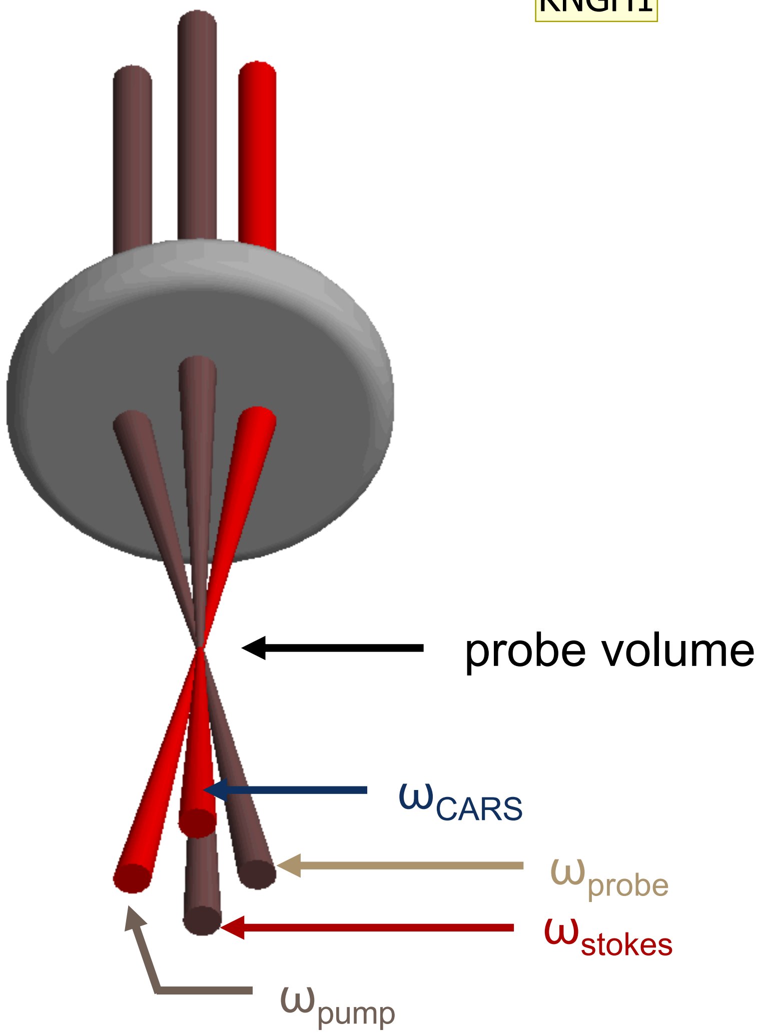
Measurement Challenges

- "Dirty" environments
  - Fire research
  - Energetic materials
- Solid particles
  - Soot
  - Aluminum
- Luminosity
- Scattering
- Absorption/optical thickness
- Large-scale of combustion systems



Coherent anti-Stokes Raman Scattering (CARS)

KNGH1



- Coherent laser-like signal beam
  - Spatially isolated
  - Readily coupled to fibers
- Blue-shifted signal beam
  - Spectrally isolated
- Orders of magnitude stronger than incoherent scattering

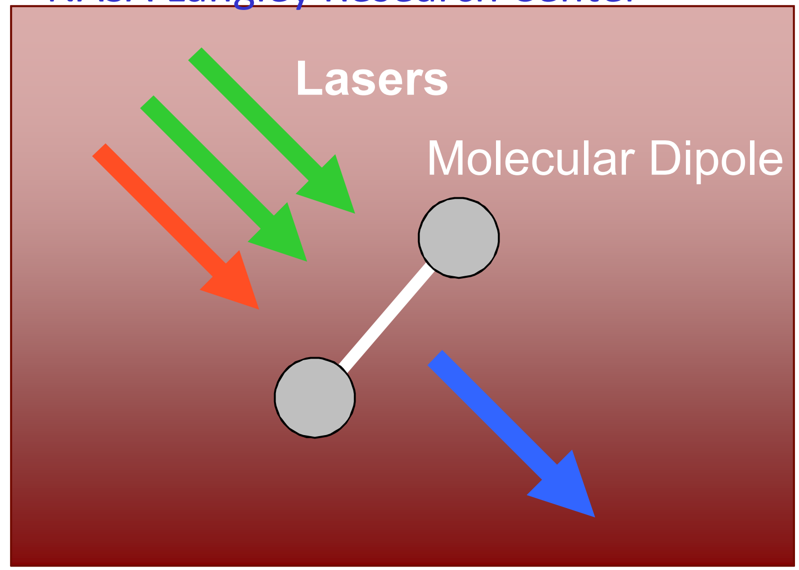
Slide 6

KNGH1 Rotate this image so labels will work better
Hoffmeister, Kathryn N Gabet, 12/16/2015

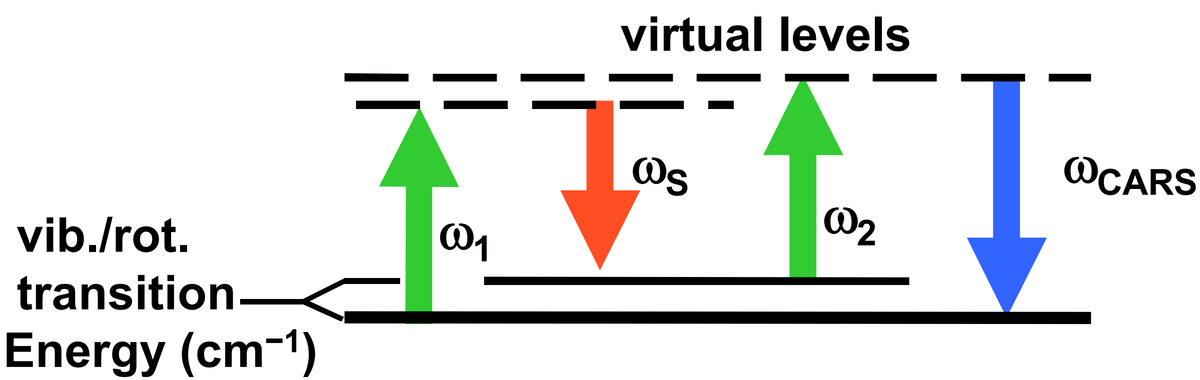
Vibrational CARS Process

- A ***'polarization'*** or induced dipole is prepared by pump and Stokes beams
- This ***polarization*** scatters the second pump wave
- Constructive interference in one ***phase-matched*** direction only

P. Danehy and S. Tedder
NASA Langley Research Center



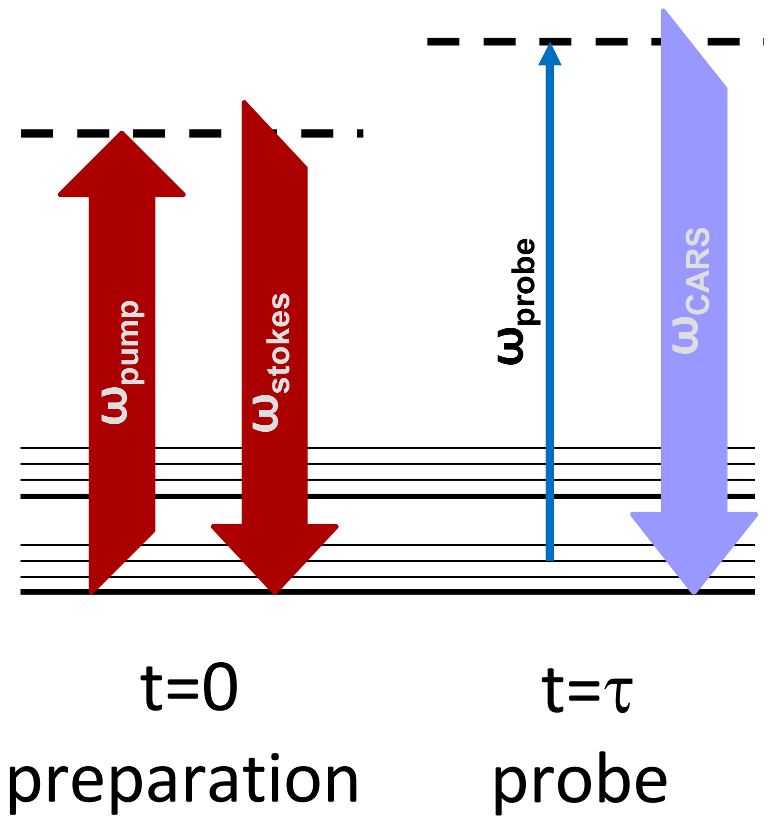
Coherent Anti-Stokes Raman



Time Domain – fs **Rotational** CARS

"The story here is really in the time domain"

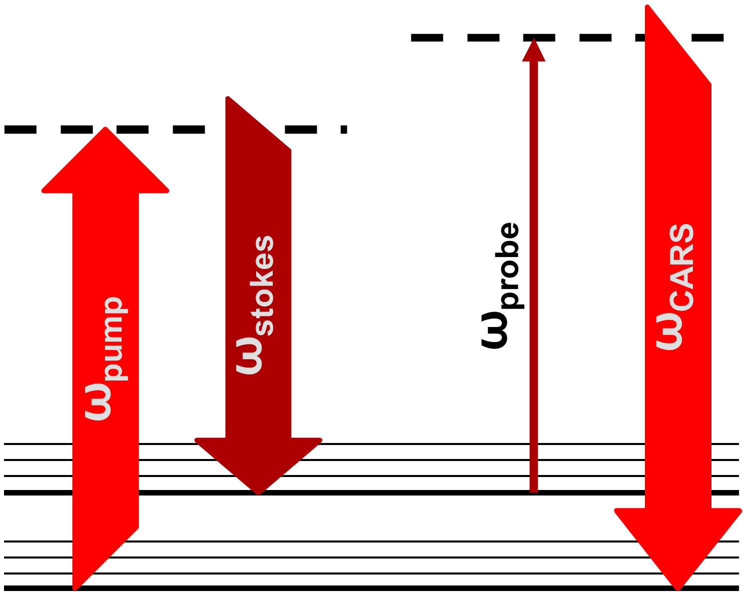
$\tau_{laser} << \tau_{molecule}$



Time Domain – fs **Vibrational** CARS

“*The story here is really in the time domain*”

$\tau_{laser} << \tau_{molecule}$

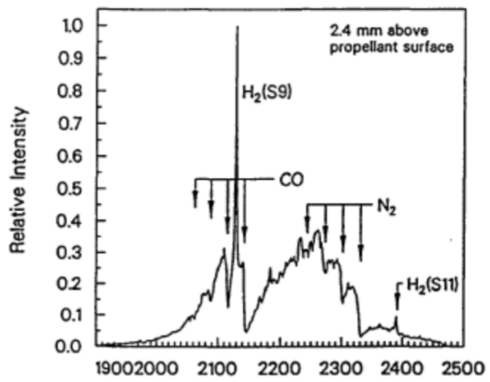


t=0
preparation

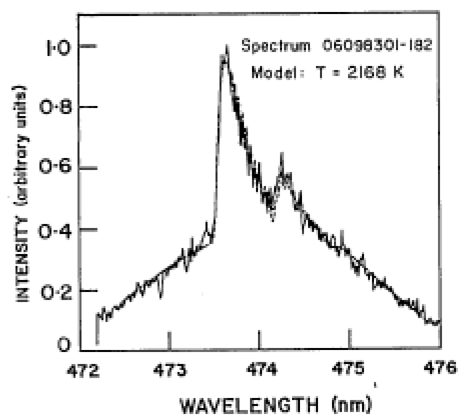
t=τ
probe

CARS Measurements in Propellants

- Conventional ns CARS measurements have been used in **non-metalized** propellants
  - RDX (Aron and Harris)
  - SGP-38 (Harris and McIlwain)
  - Nitramines (Harris; Stufflebeam and Eckbreth)
  - HNF (Dragomir *et al.*)
- Nonresonant background a problem at high pressures
- Closest thing to Metalized propellants is coal combustion
  - Beiting; Hancock; Lucht



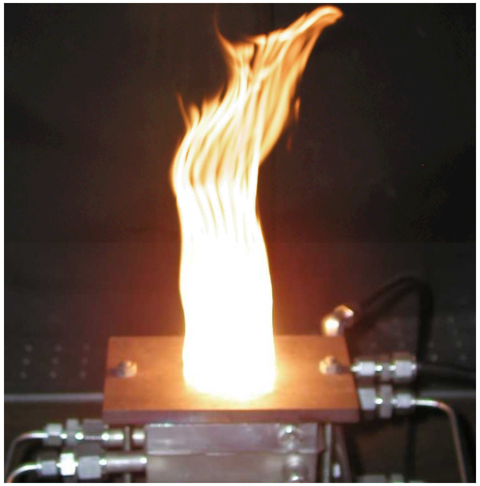
ns-CARS spectrum HMX/TMETN at high pressure Stufflebeam and Eckbreth

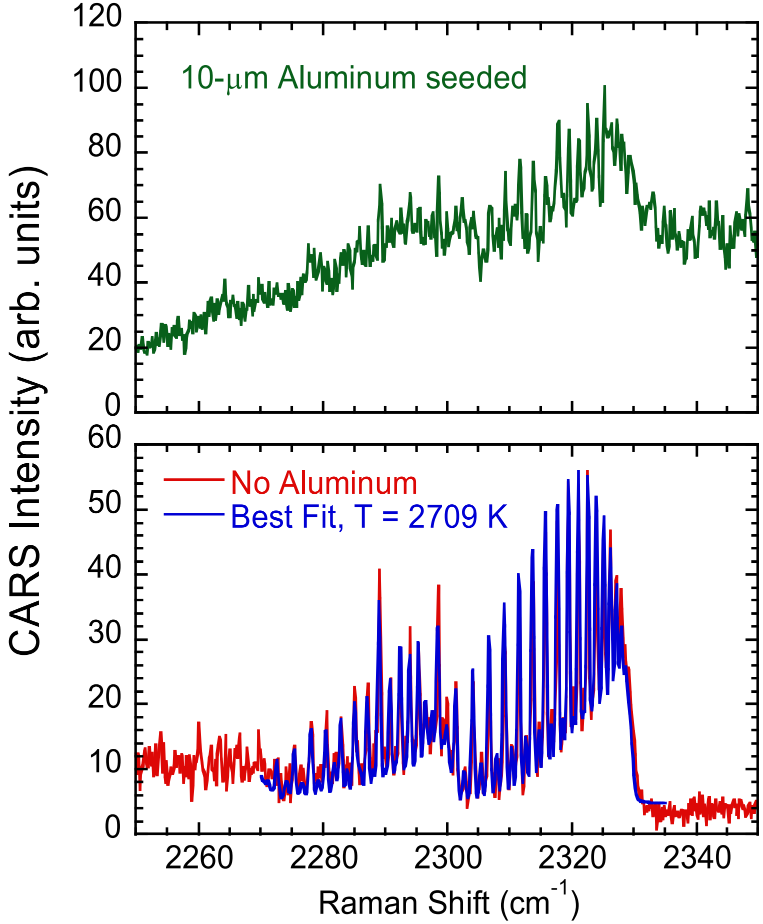


ns-CARS spectrum coal-fired MHD environment Beiting

ns Vibrational CARS Spectra

- ns vibrational CARS spectra taken in acetylene/air flames
  - With and without aluminum particle seeding
- Nonresonant background problem in seeded flames

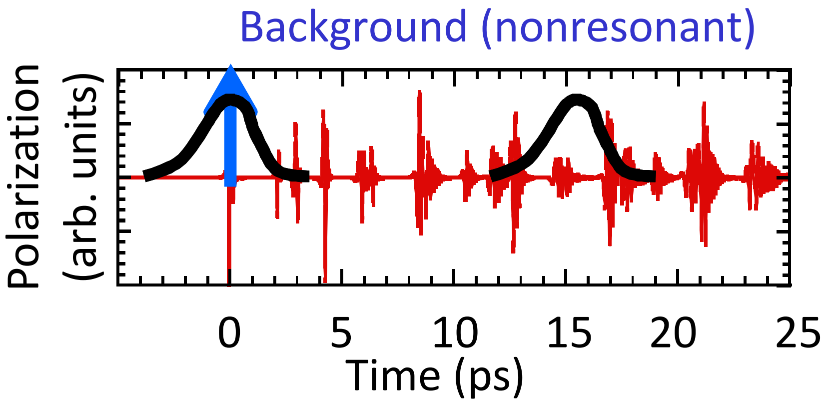


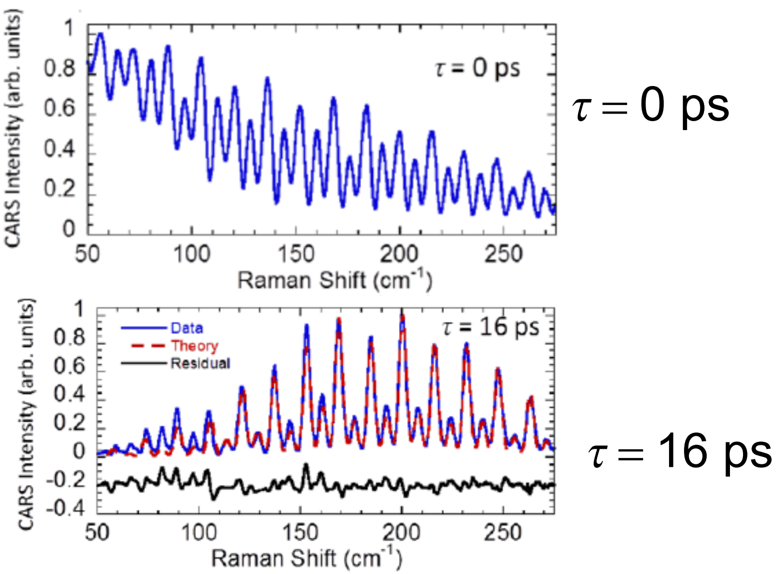


C₂H₂/air flame

Advantages to Ultra-fast CARS

- Low total pulse energy
  - ~1 mJ or less (factor of ~40-100 lower than ns laser pulses)
  - Reduces likelihood of breakdown-like interference
- Time-gate elimination of nonresonant background
- Raman-resonant signal results from much slower, nuclear response of the molecule





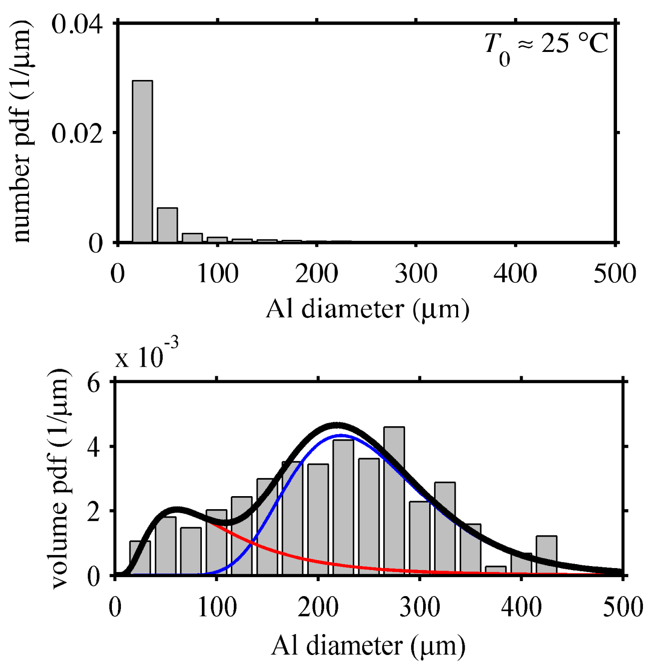
AP Propellant Strand Burns

- Aluminized ammonium-perchlorate (AP) propellant strands
- Acquired ~60,000 single-shot spectra over strand-burner duration
- Hostile environment for diagnostics
  - Very high temperatures – fluctuations over 3,000K
  - Dense field of large molten-metal particles
- Significant number of spectra rejected, but over 20,000 were retained for valid temperature measurements

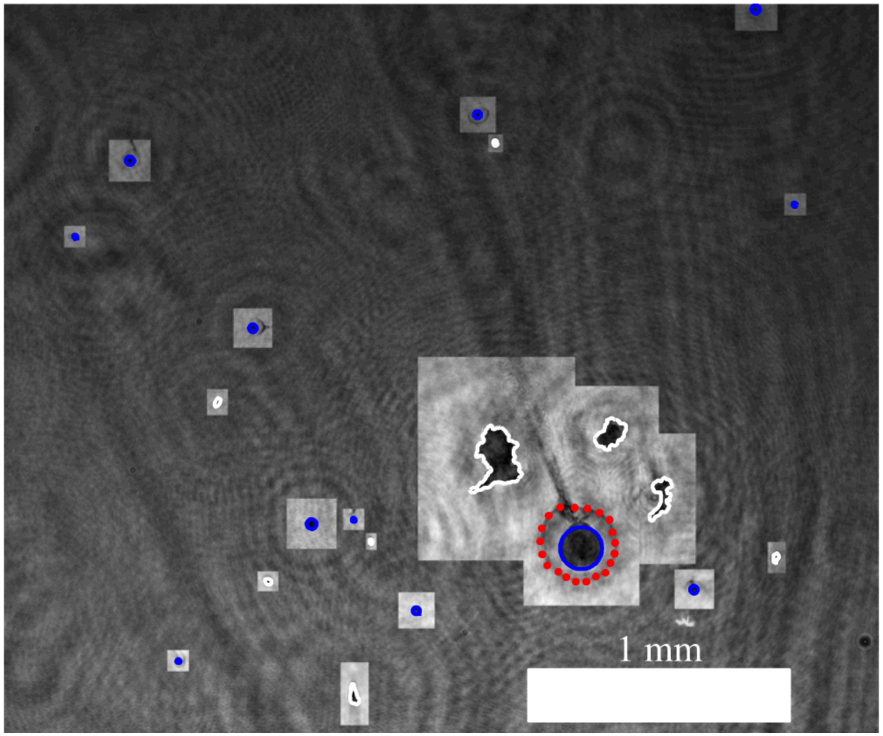
Video Recorded During CARS Measurements

AP Propellant Strand Burns (cont)

- Particle sizes measured with Digital In-line Holography (DIH)
  - From Guildenbecher *et al.*



<u>*Al Particle Size Distributions*</u>



<u>*Refocused DIH hologram*</u>

fs/ps SHBC CARS Instrument

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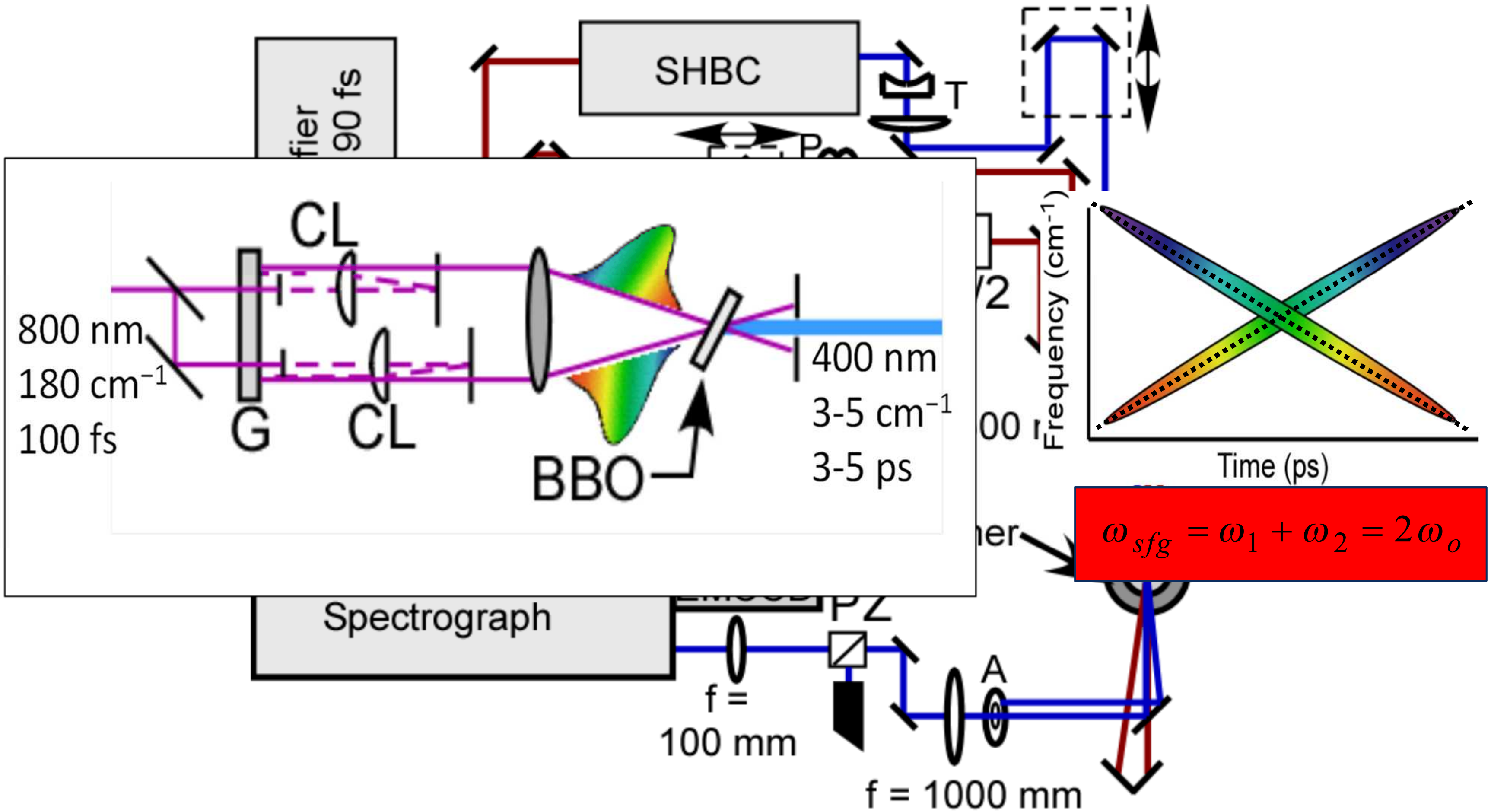
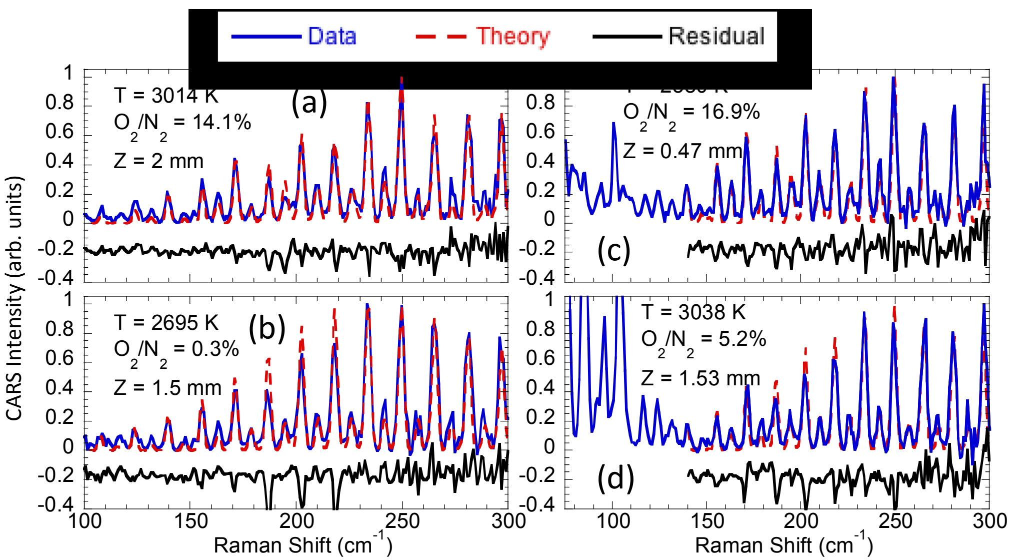
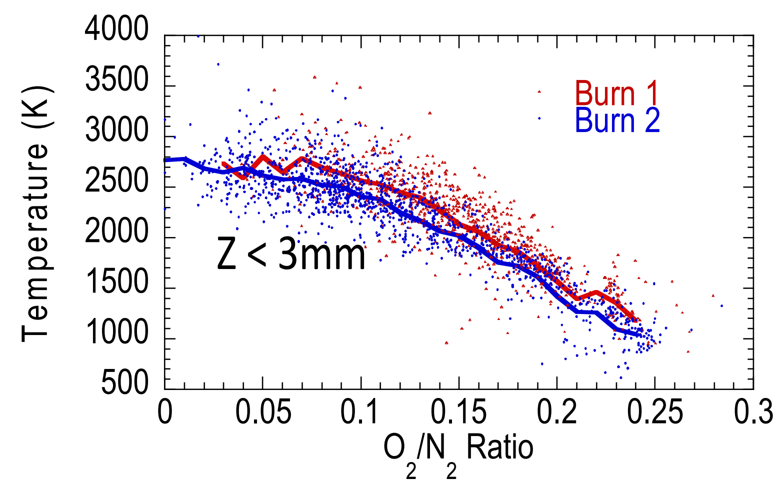


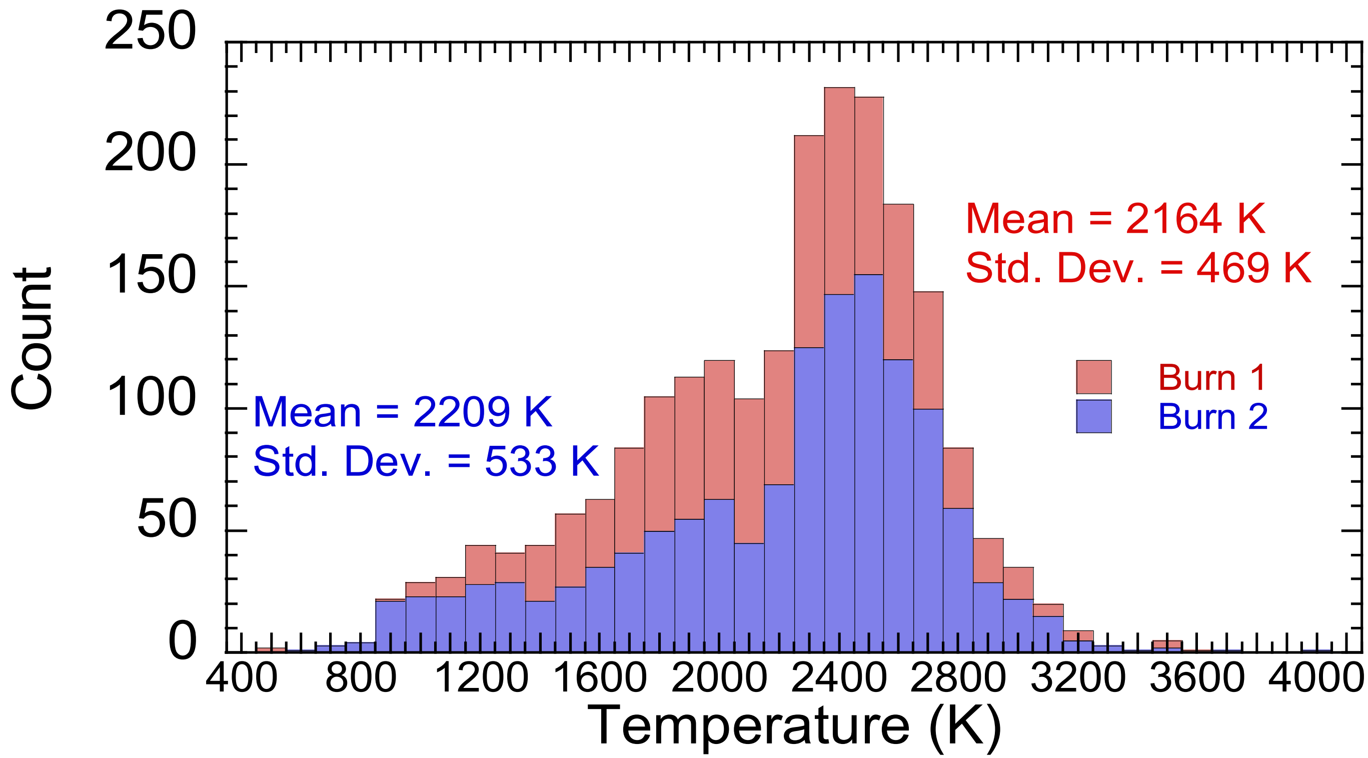
CARS Results in Propellant Burn

- Results for CARS measurements within 3 mm of propellant surface shown
  - Height estimated from 1-kHz acquisition rate and burn rate of 800 µm/sec
- Spectra dominated by N_2 and O_2 contributions
  - Cl_2, HCl, NO, NO_2 also present





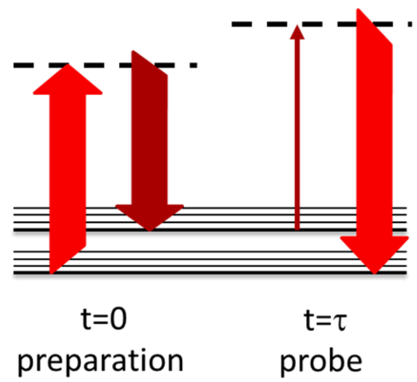
CARS Results in Propellant Burn

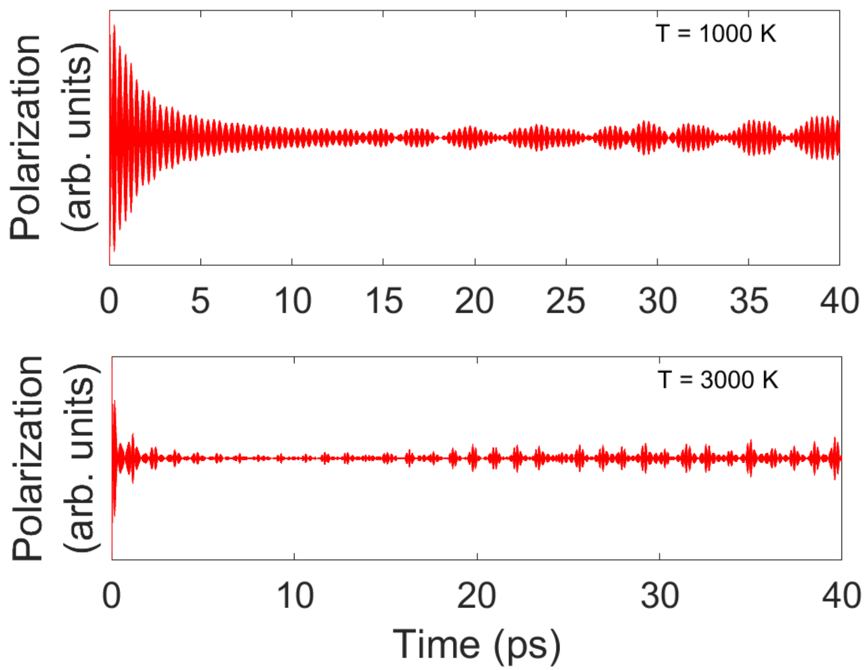


Switch to vibrational CARS for follow-on experiments

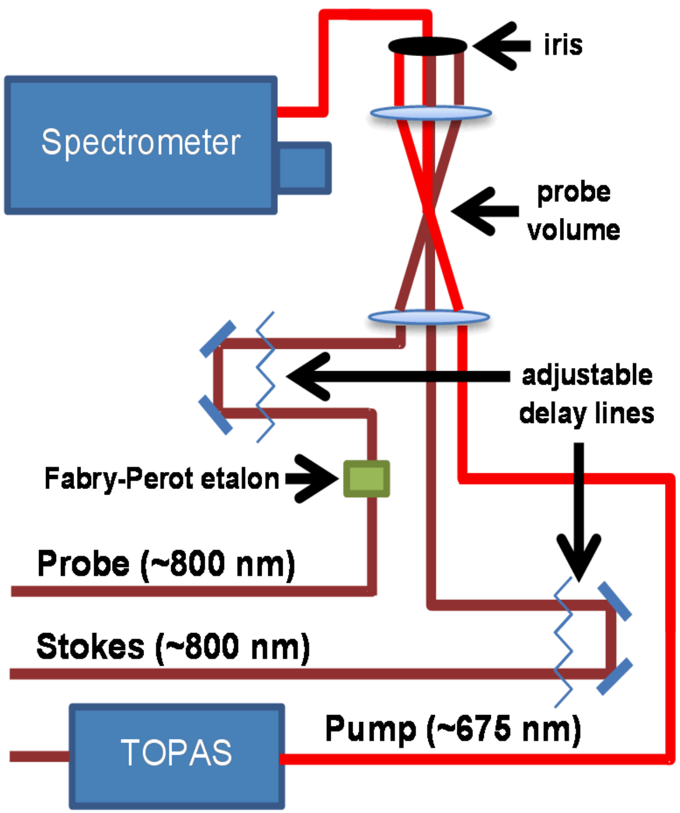
Vibrational CARS

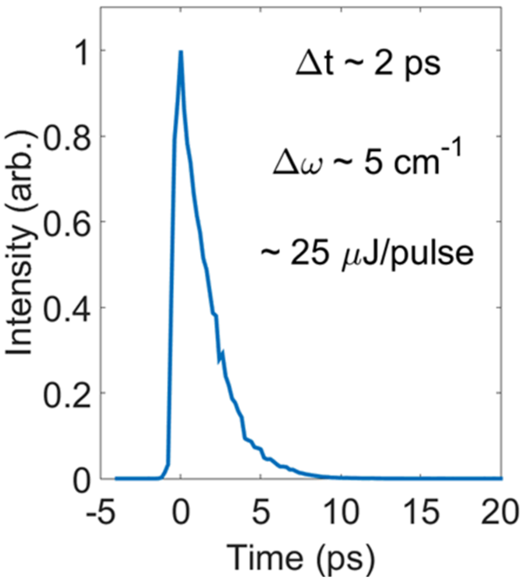
- Want to acquire data away from non-resonant background at (τ=0 ps)
- Resonant coherence decreases with time, but increases again at long time delays (Miller *et. al*, JRS 2015)
- Spectral shape is also important





Hybrid Vibrational CARS Instrument

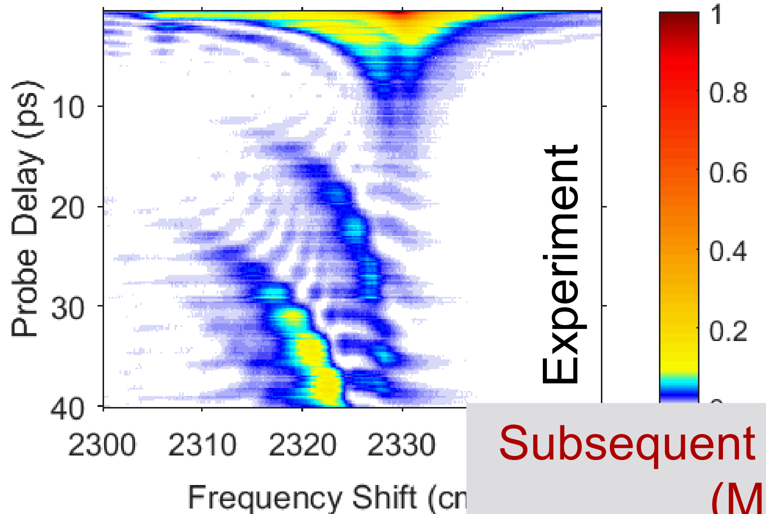




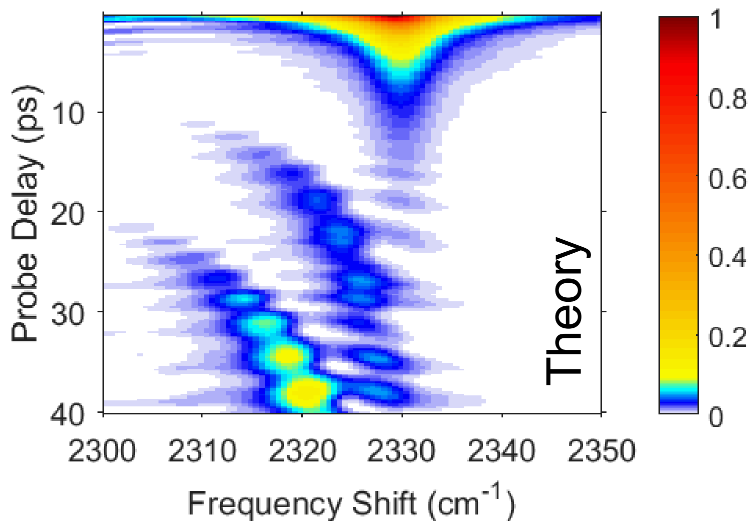
Probe Beam from etalon

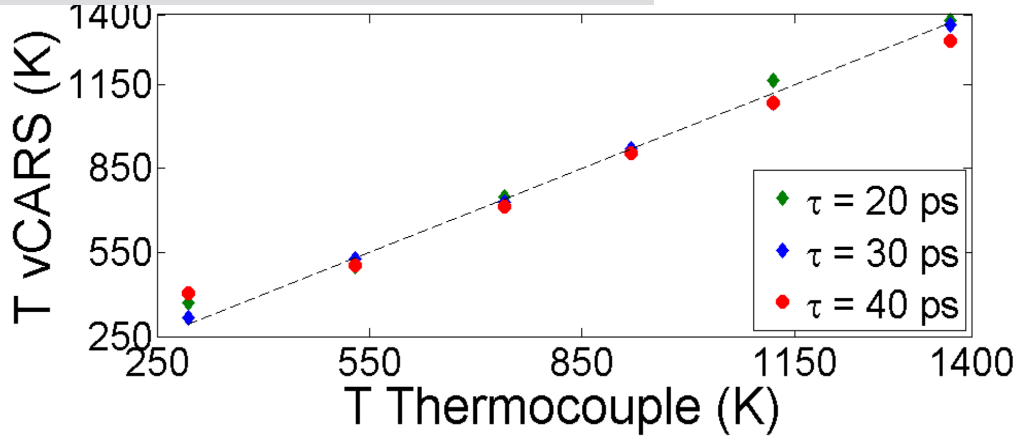
Furnace Measurements

- Measured temperature at time delays varying from τ = 0 to 40 ps
- Fit data taken in furnace at T = 300 to 1300 K



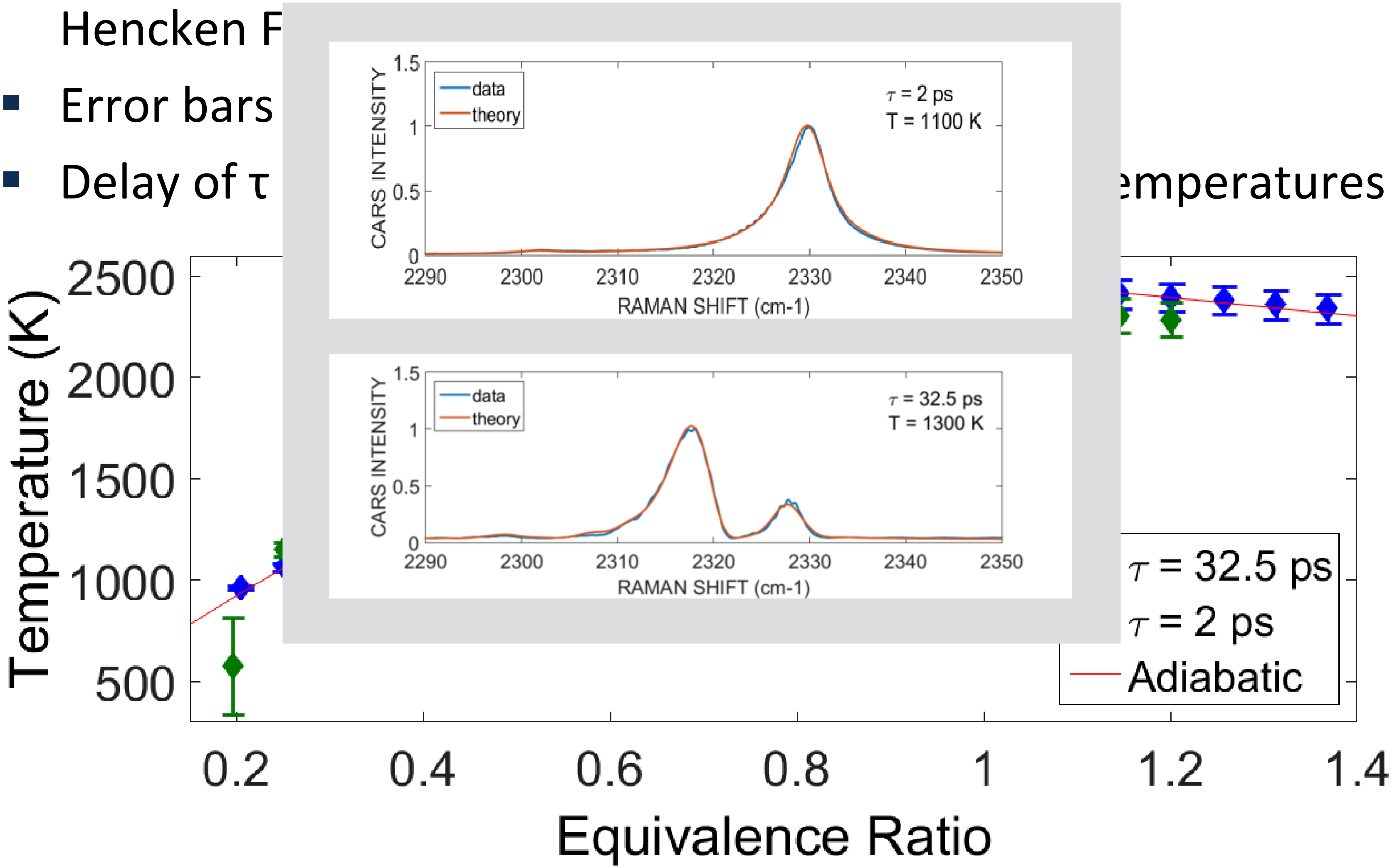
Subsequent data taken at τ = 2 and 32.5 ps
(Miller *et. al*, JRS 2015)





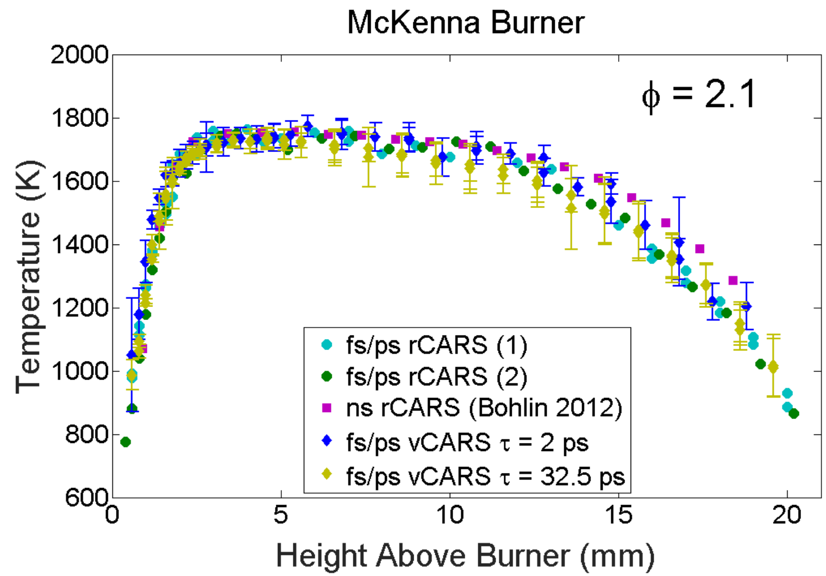
Clean, Near-Adiabatic Flame

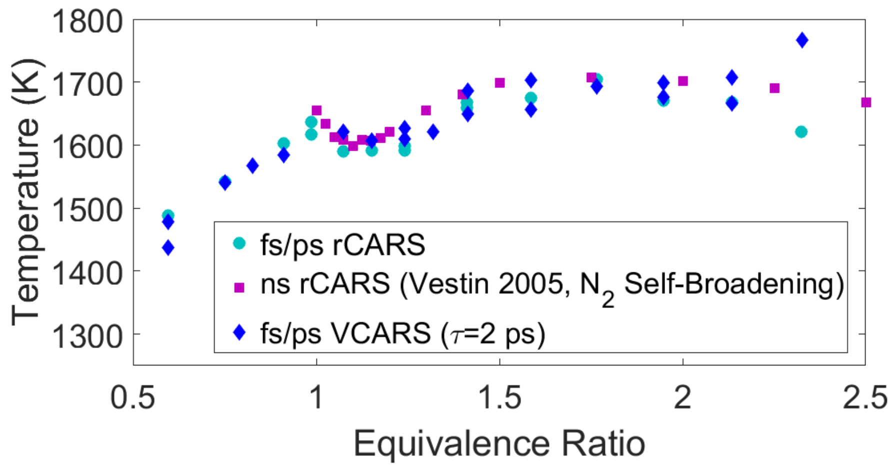
- Results for vibrational CARS measurements in near-adiabatic Hencken F
- Error bars
- Delay of τ … emperatures



Sooting Flat-Flame Burner

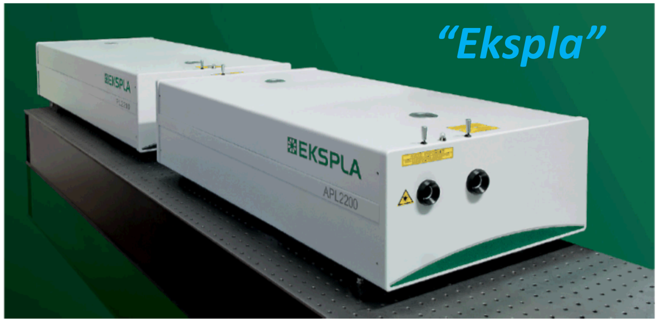
- Measured temperature at in a ethylene-air Flame (McKenna Burner)
- LII Workshop conditions
  - Soot inception low in flame
  - Up to ~75 ppb soot near stabilization plate in ϕ = 2.1 flame (Hadef *et al.*, 2010)
  - Up to ~70 ppb soot at 8.5 mm in ϕ = 2.5 flame (Hadef *et al.*, 2010)
- First hybrid vibrational CARS measurements in highly-sooting flames



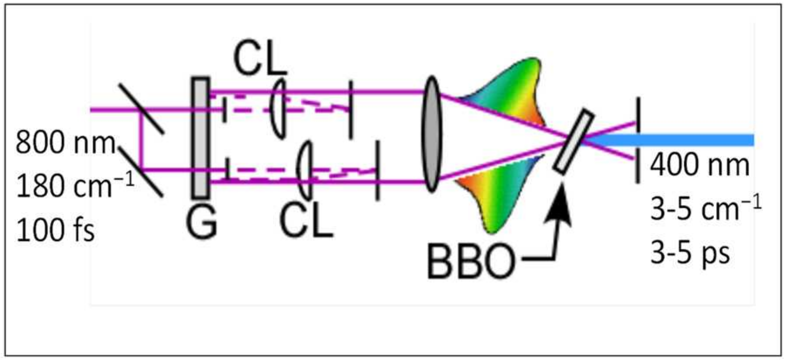


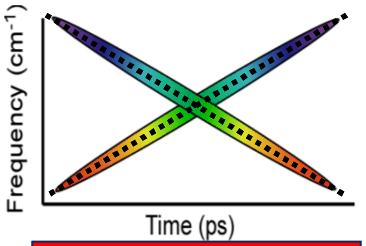
Future Work

- Application of hybrid Vibrational CARS system to propellant measurements by increasing probe energy
  - ps Nd:YAG regenerative amplifier (Bohlin *et al.*, 2013)



  - Second-Harmonic Bandwidth Compressor





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

Conclusions

- Measured temperatures in aluminized ammonium-perchlorate (AP) propellant strands
  - Utilizing hybrid fs/ps rotational CARS instrument
  - Spectra dominated by N_2 and O_2 contributions
  - Cl_2, HCl, NO, NO_2 also present
  - Measured temperatures vary by 3000 K
- Developed a new hybrid fs/ps vibrational CARS instrument
  - Validated code to predict spectral response
  - Quantified precision in furnace and canonical flat-flame environments
  - Applied technique in sooting flame (LII workshop, McKenna burner)

Acknowledgements

- Thanks again to Howard Lee Stauffacher for his handling and setup of the propellant strands.
- Sandia is a multirogram laboratory operated by Sandia Corporation, a Lockheed-Martin Company, for the United States Department of Energy's National Nuclear Security Administration under Contract DE-AC04-94AL85000.

Questions?

Questions?