Coherent anti-Stokes Raman Scattering for Determining Key Thermodynamic Properties of Supersonic, Combusting, and Nonequilibrium Flows

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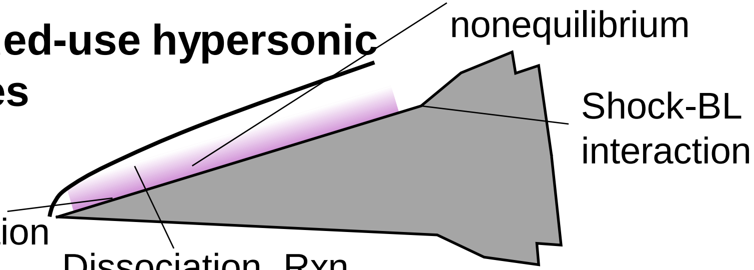
Coherent anti-Stokes Raman Scattering

Flowfields of interest characterized by:

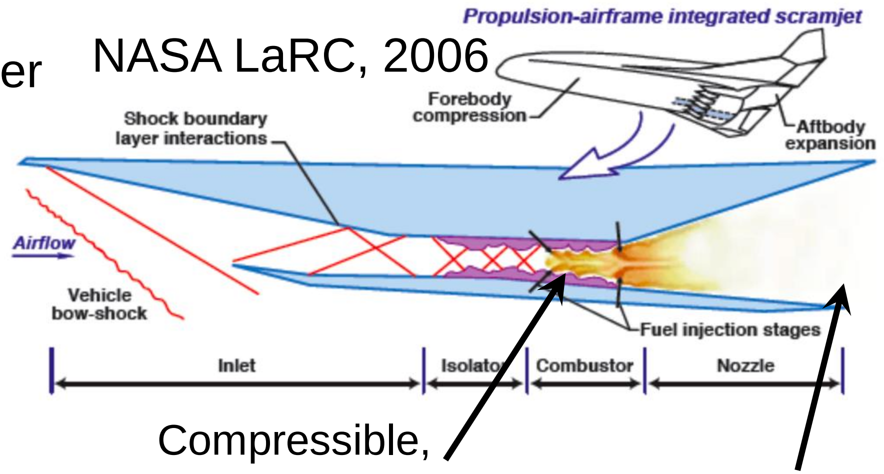
- Reactions (combustion, dissociation/ recombination, etc.)
- Nonequilibrium energy distributions and resulting reactions, internal energy transfer
- Wide range in temperatures, pressures
- Spatial gradients
- Transient flow dynamics

Extended-use hypersonic vehicles



N_2, O_2, O, N, NO, O_2^+, N_2^+

High-speed vehicle engines (air-breathing, rocket), exhaust plumes



NASA LaRC, 2006

Compressible, turbulent combustion

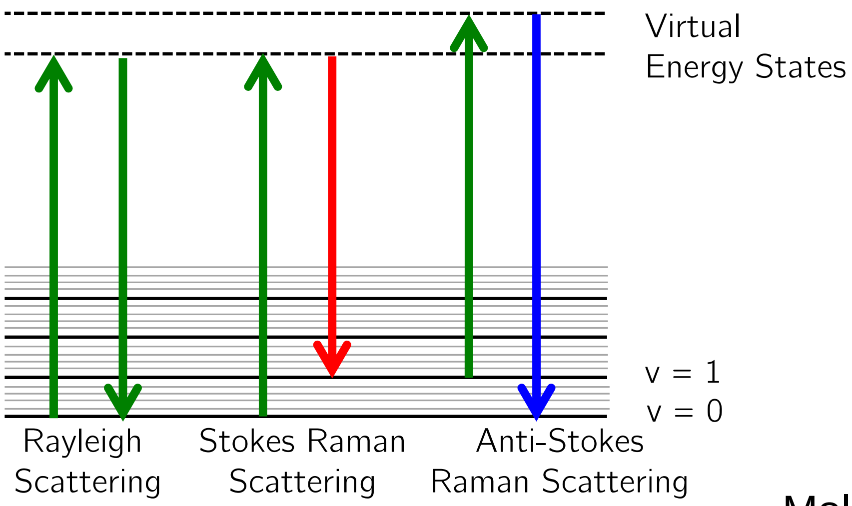
High-energy, radiative exhuast

***Goal**: employ fs/ps CARS to quantify thermodynamic properties with spatial resolution in highly transient, reacting flows using ground-based experimental facilities*

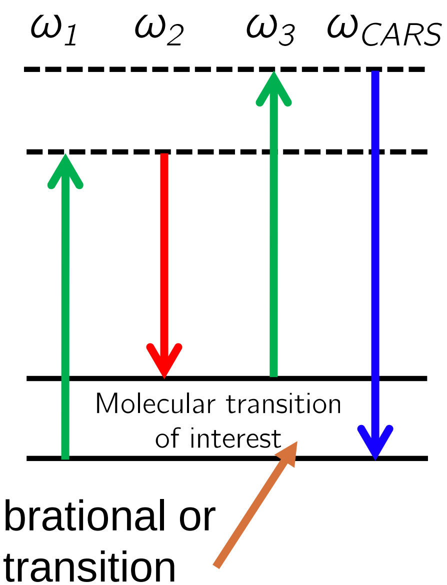
Coherent anti-Stokes Raman Scattering

Instead of relying on a change in polarizability from a naturally occurring vibration/rotation, we can drive the change in polarizability (*drive the coherence*) using two electric fields to increase strength of transition, scatter from this prepared coherence with a third field.

Raman Scattering



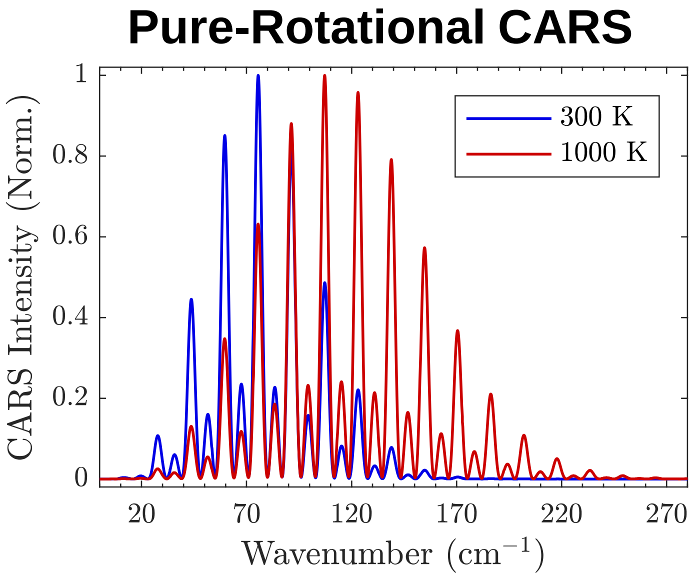
CARS

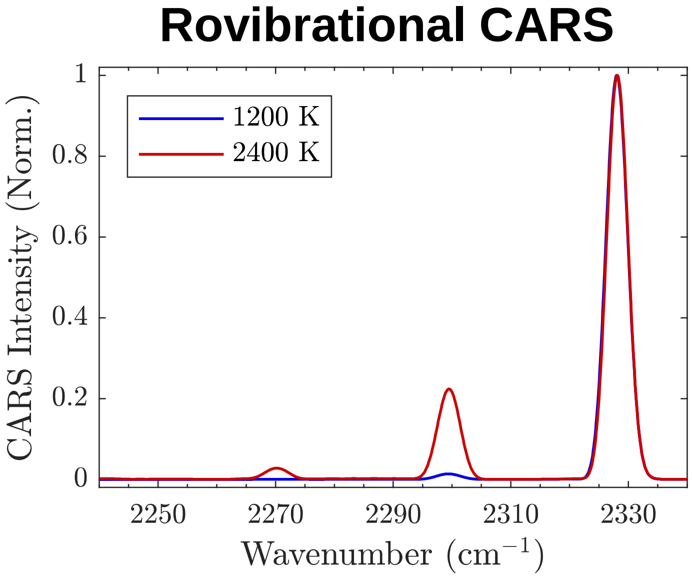


Molecular vibrational or rotational transition

Thus, three waves incident on a medium, medium response (phased array of oscillators) produces a fourth wave radiated by the induced nonlinear polarization

Temperature Sensitivity


Pure-Rotational CARS


Rovibrational CARS

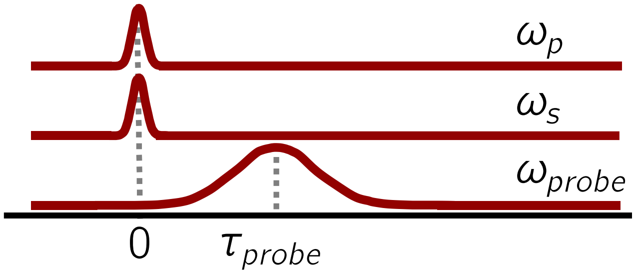
Temperature sensitivity

$$\Delta\rho_{i\rightarrow f} = \frac{g_f}{\sum_m g_m e^{\frac{-E_m}{k_B T}}}\left[e^{\frac{-E_i}{k_B T}} - e^{\frac{-E_f}{k_B T}}\right]$$

Coherent anti-Stokes Raman Scattering

Advantages of hybrid fs/ps CARS

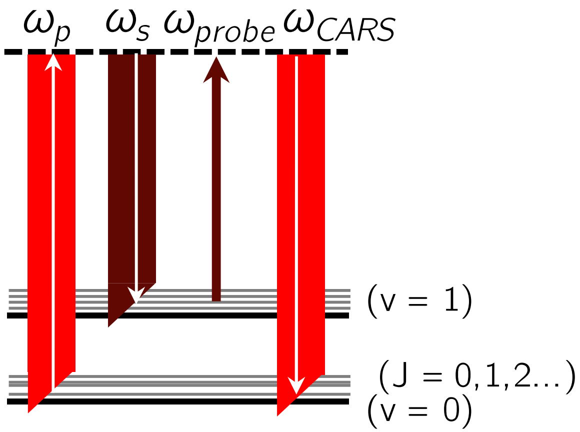
- High signal levels (highly-efficient generation of Raman coherence w/ fs pulses)
- Suppress nonresonant signal
- Insensitive (or *selectively* sensitive) to collisions
- Inherent broadband excitation: excite multiple transitions with multiple photon pairs
- Extension to 1D- and 2D-measurements (Bohlin et al., 2013; Miller et al. 2016.; Bohlin et al. 2017)



Pulse Timing

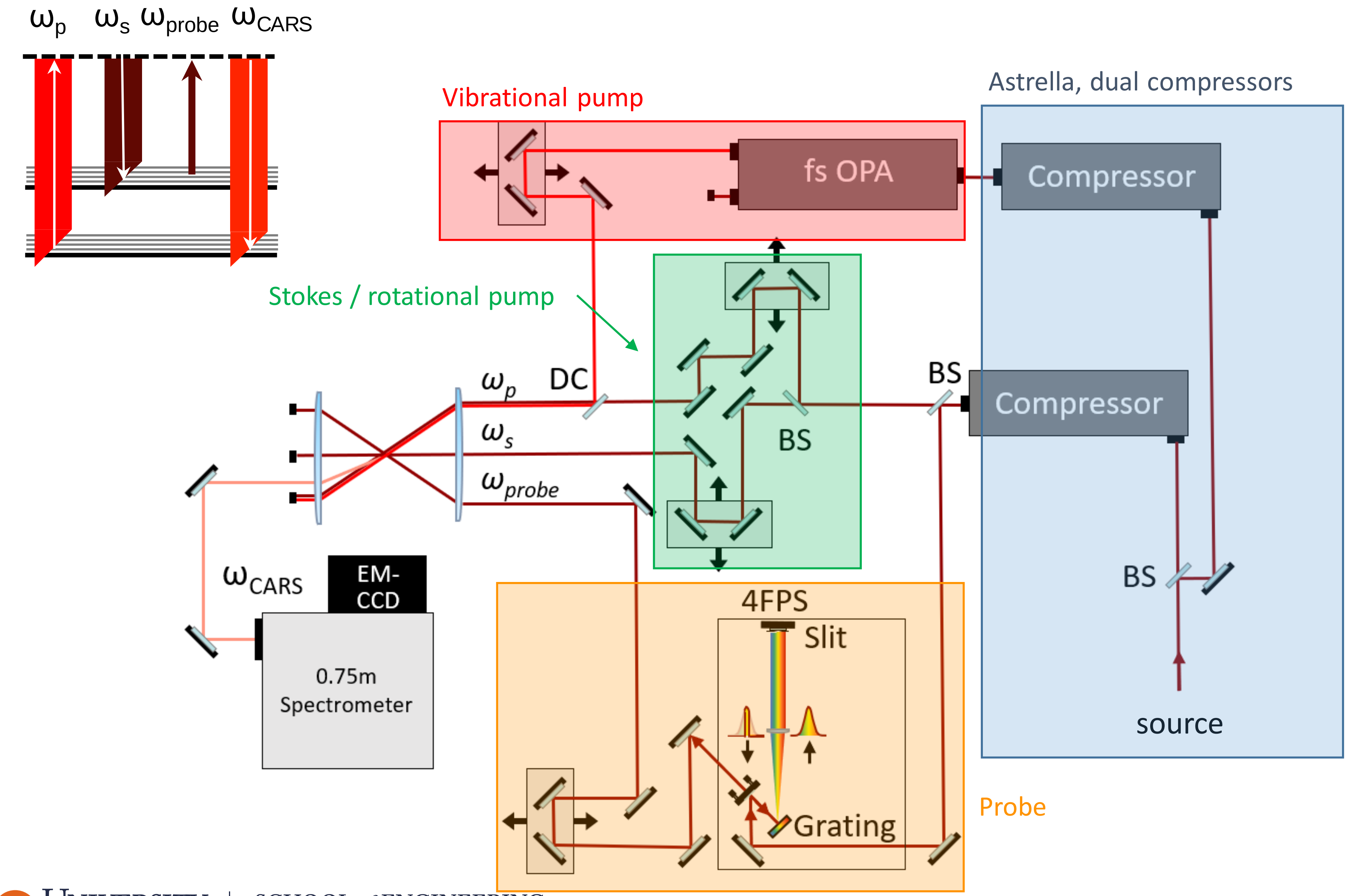
Transform-limited

$$I_{CARS}(t) \xleftrightarrow{\mathcal{F}} I_{CARS}(\omega)$$



Frequency Diagram

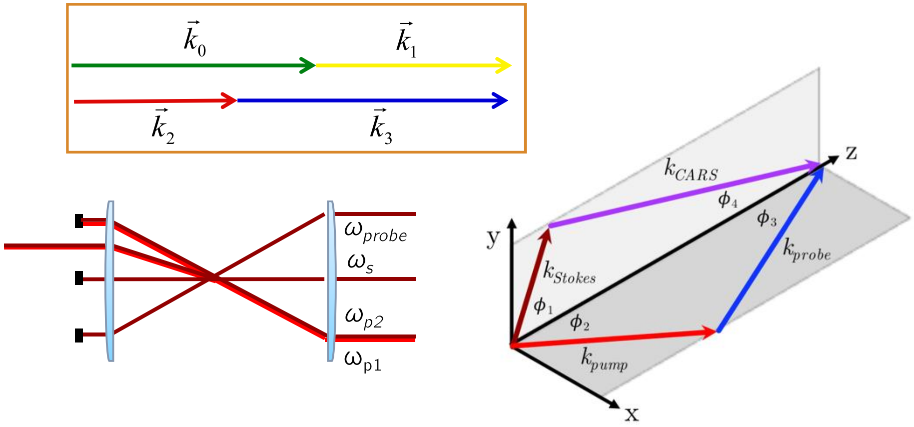
Coherent anti-Stokes Raman Scattering



Spatially-resolved measurements

Required spatial and temporal overlap of the 3 incoming electric fields can provide a spatially-resolved measurement, depending on phase-matching configuration.

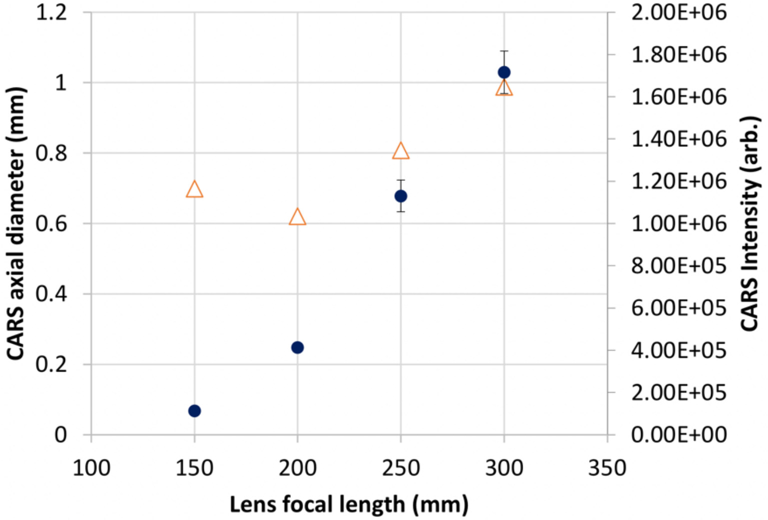
Phase-Matching Configurations



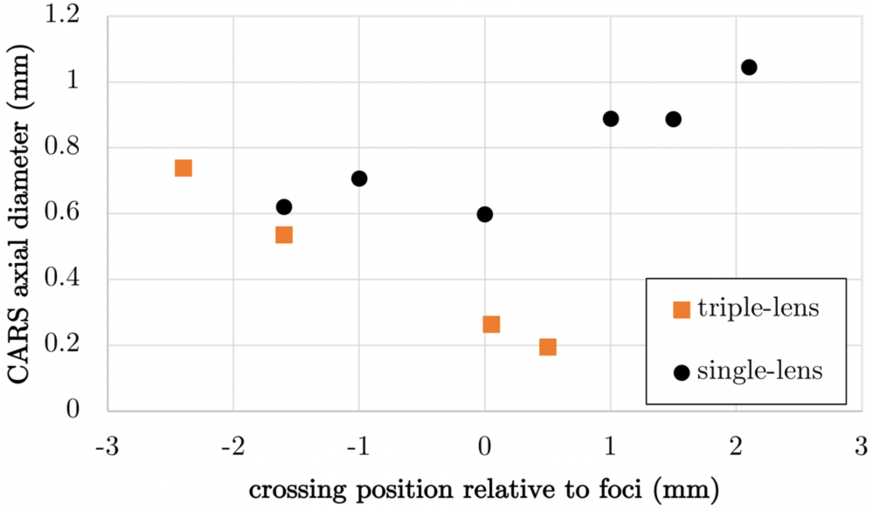
$$E(\mathbf{r},t) = E(t)e^{i(\mathbf{kr}-\omega_0 t+\phi)}$$

$$\vec{k}_1 + \vec{k}_2 - \vec{k}_3 - \vec{k}_4 = 0$$

Signal intensity vs. resolution

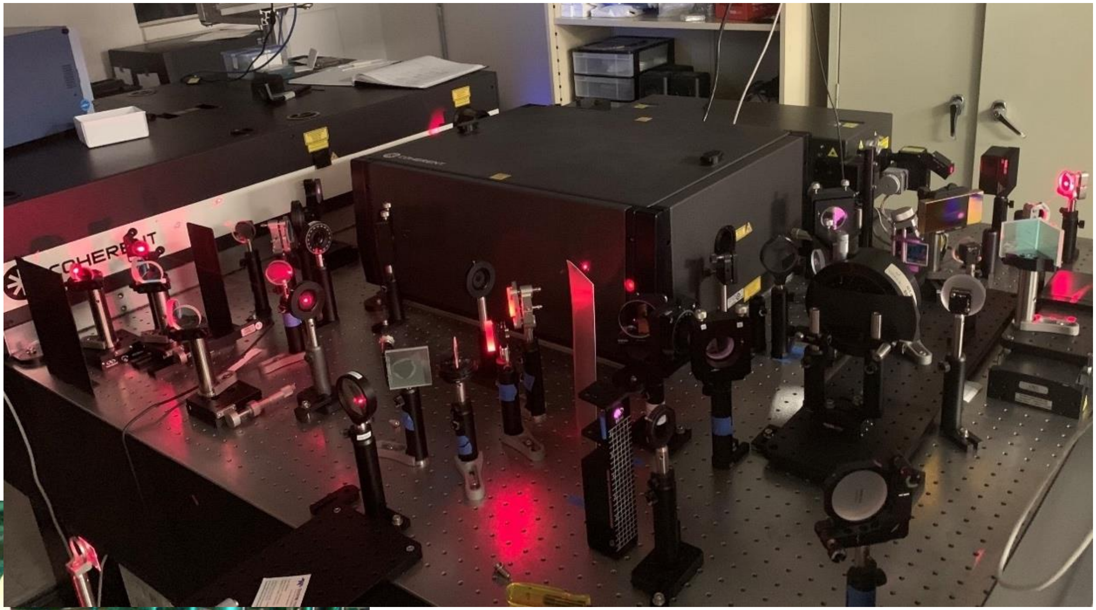


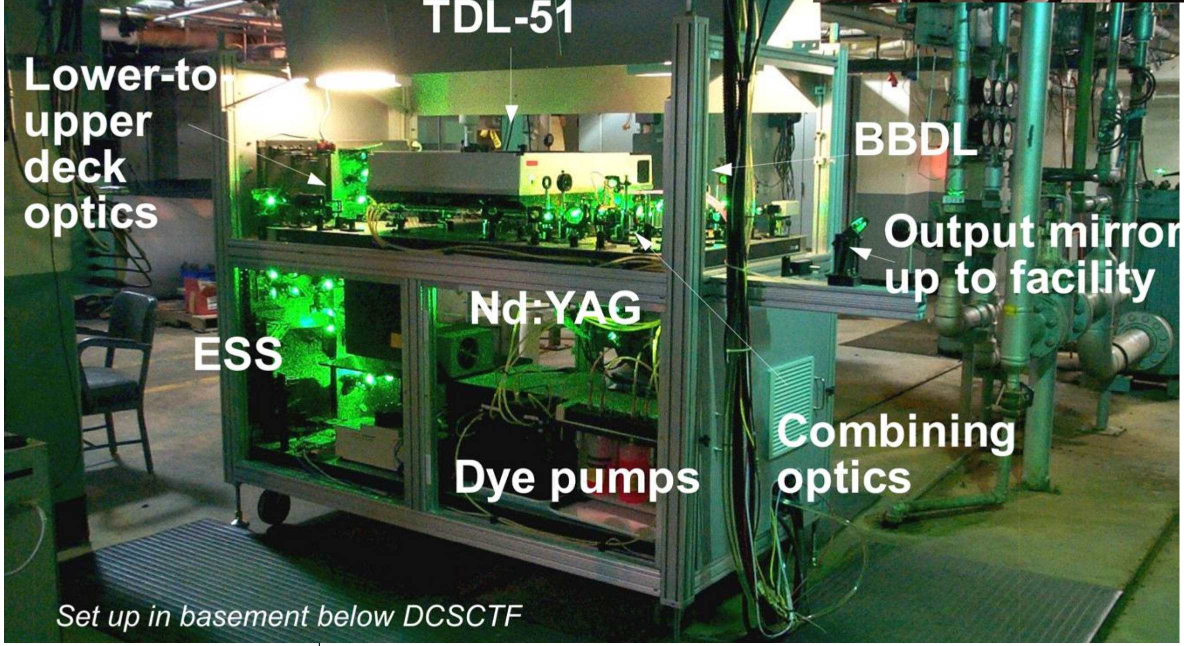
Optimizing resolution



CARS systems

UVA fs/ps CARS system
(C. Dedic)





Set up in basement below DCSCTF

GWU ns CARS cart (A. Cutler)

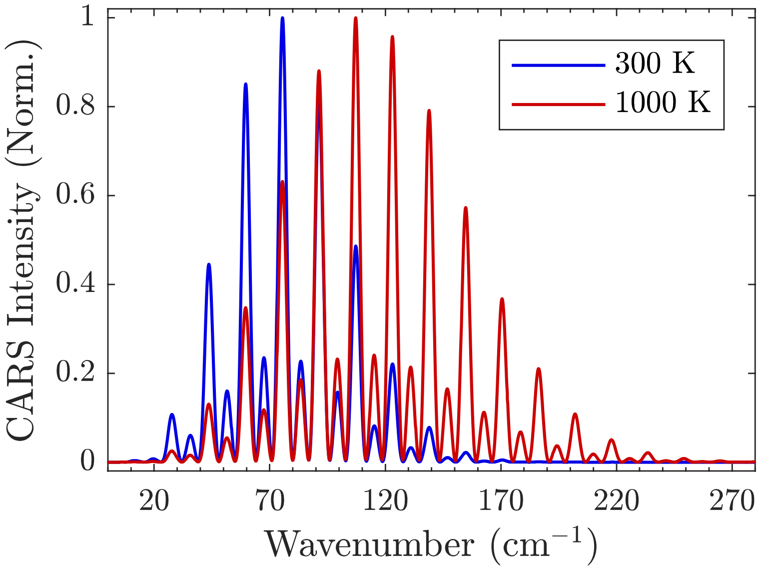
P. M. Danehy, B. F. Bathel, C. Johansen, M. Winter, A. D. Cutler, S. O'Byrne, "Molecular-based optical measurement techniques for Nonequilibrium Hypersonic Flows," book chapter in AIAA Progress Series Book, Vol. 247, entitled "Hypersonic Nonequilibrium Flows", edited by Eswar Josyula, published May, 2015.

Temperature

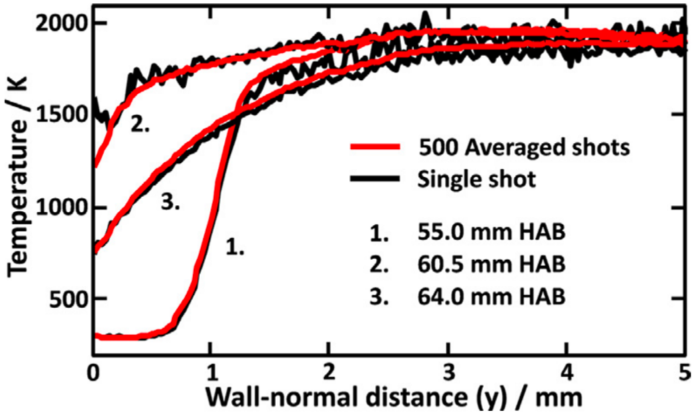
Experimentally determined error/precision in single-shot temperature evaluations for…

	Accuracy	*Precision*	
ns CARS	2-5%	3-5%	Seeger, Leipertz, *Appl. Opt.*, 1996
fs/ps CARS	1-5%	1-3%	Miller et al., *Opt. Lett.*, 2010 Miller et al., *Opt. Exp.,* 2011
	<3%	<2%	Kearney, *Combust. Flame*, 2015

Temperature-sensitive response:

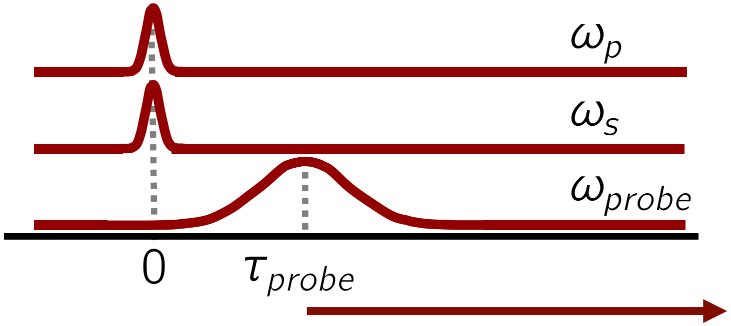


Extension to 1D measurements:

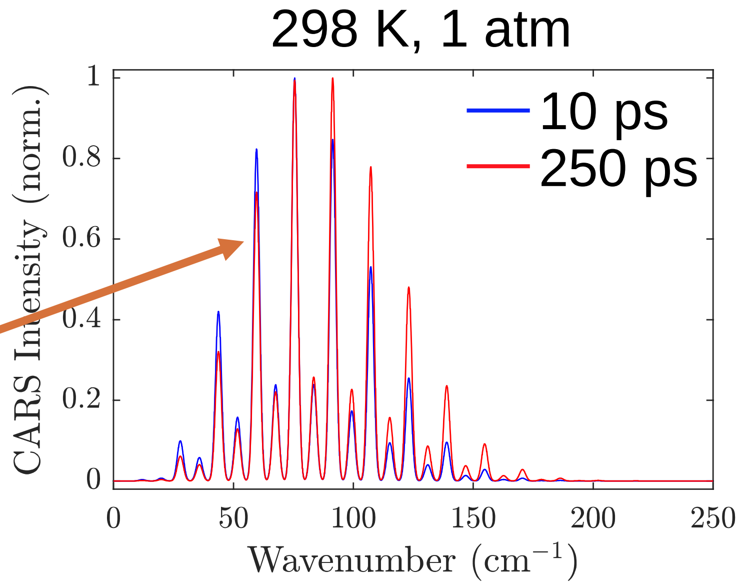


Bohlin, Jainski, Patterson, Dreizler, Kliewer, **Proc. Combust. Inst., 2017**

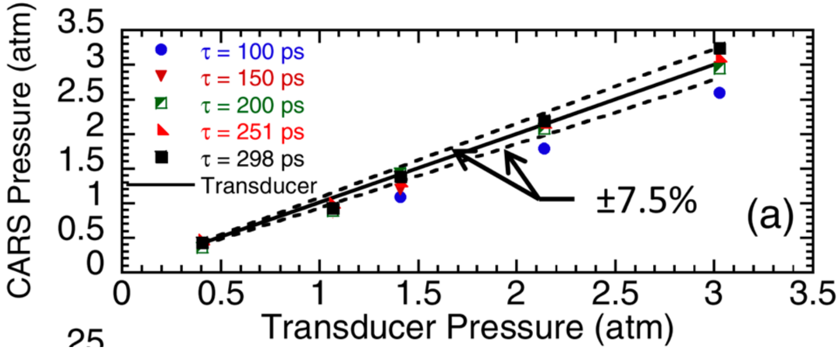
Pressure



Delay probe pulse to sample the **time response** of Raman excitation

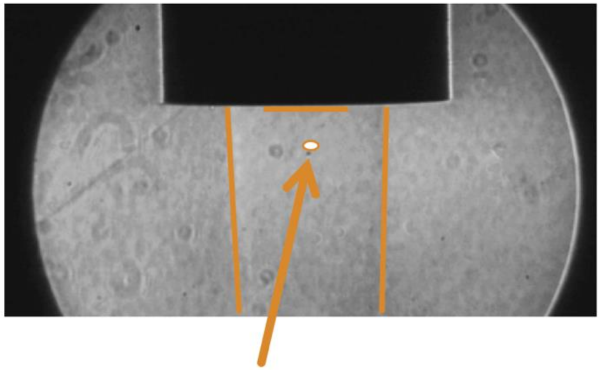


Measure the *relative* decay of each transition to determine pressure from collisional-induced signal decay

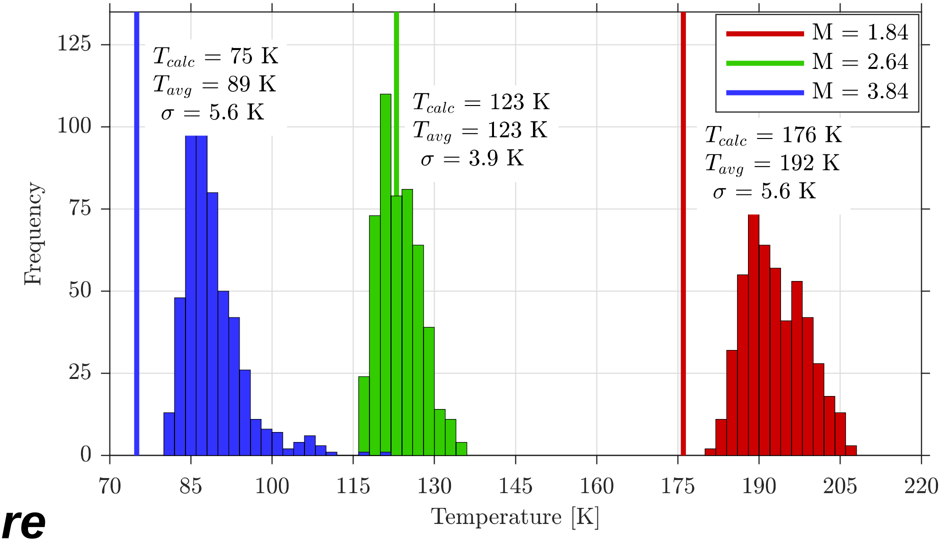


Kearney and Danehy, AIAA SciTech, 2015
Kearney and Danehy, Opt. Lett., 2015

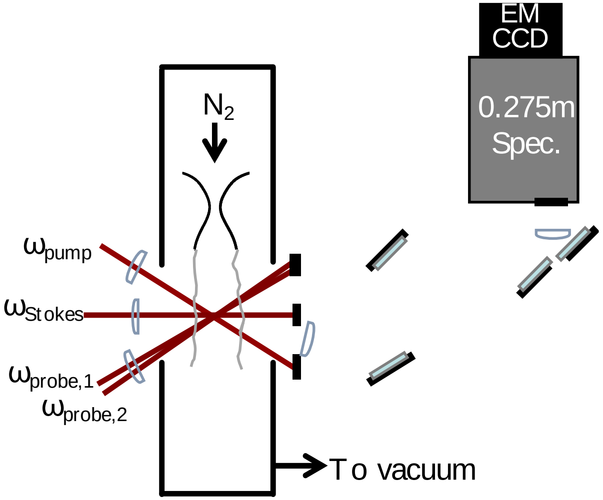
Pressure



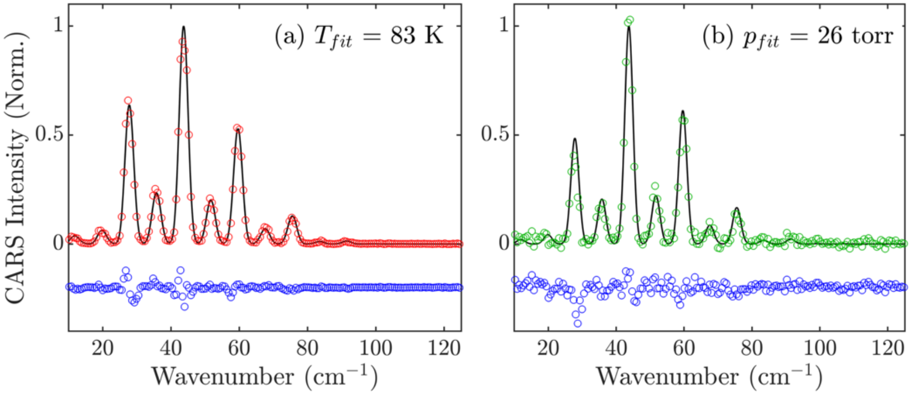
1mm CARS probe volume



*2-probe technique to measure **temperature** and **pressure** fluctuations in a supersonic jet*



T_{fit} = **83 K** p_{fit} = **26 torr**

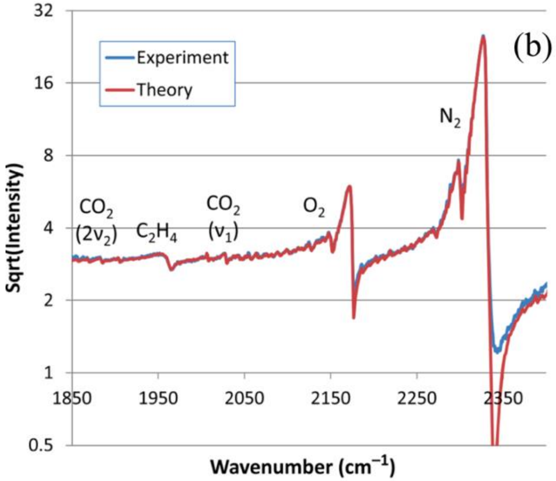


Dedic, Cutler, Danehy, AIAA SciTech 2019

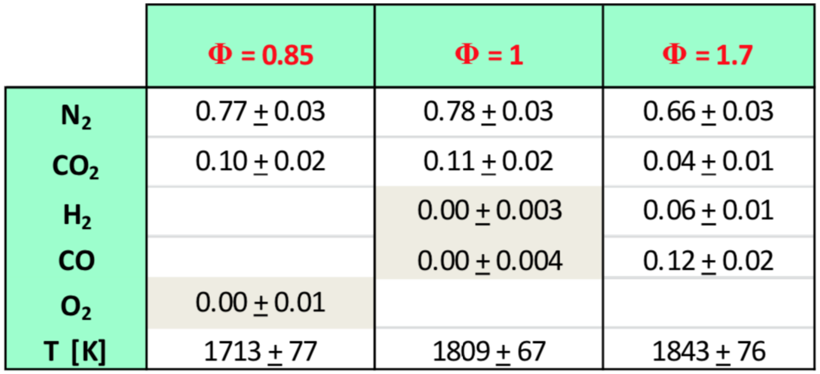
Species concentration

WIDECARS (N_2, CO_2, H_2, CO, O_2, C_2H_4)

Cutler, Gallo, Cantu, **Appl. Opt., 2017**



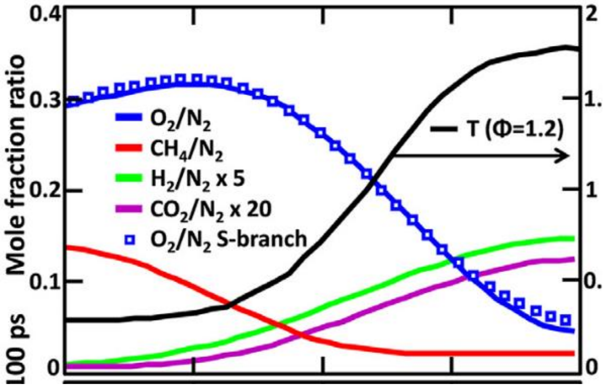
Gallo, Cantu, **Cutler**, Rahimi, Chelliah, **AIAA AVIATION, 2014**



	$\Phi = 0.85$	$\Phi = 1$	$\Phi = 1.7$
N_2	0.77 ± 0.03	0.78 ± 0.03	0.66 ± 0.03
CO_2	0.10 ± 0.02	0.11 ± 0.02	0.04 ± 0.01
H_2		0.00 ± 0.003	0.06 ± 0.01
CO		0.00 ± 0.004	0.12 ± 0.02
O_2	0.00 ± 0.01		
T [K]	1713 ± 77	1809 ± 67	1843 ± 76

fs/ps CARS w/ supercontinuum generation (N_2, CO_2, H_2, CO, O_2, CH_4)

Bohlin, Jainski, Patterson, Dreizler, Kliewer, **Proc. Combust. Inst., 2017**

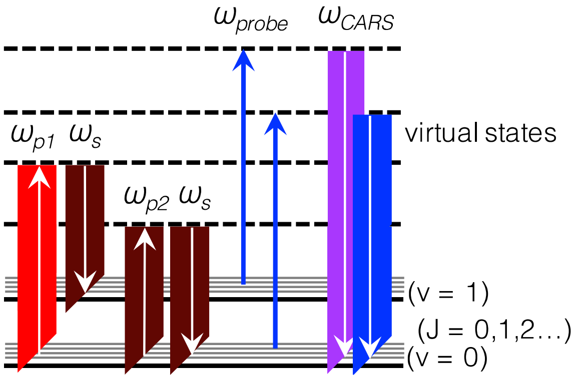


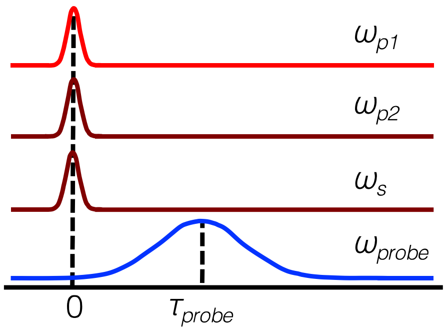
ERE CARS for minor species measurements (CH, OH, NO)

Nonequilibrium energy distributions

Hybrid fs/ps CARS is ideal for studying nonthermal environments

- → Single-shot measurement of rotational *and* vibrational energy distributions
- → Resolve spatial non-uniformities
- → Capture highly transient phenomena
- → Suitable for studying local internal energy transfer and thermalization

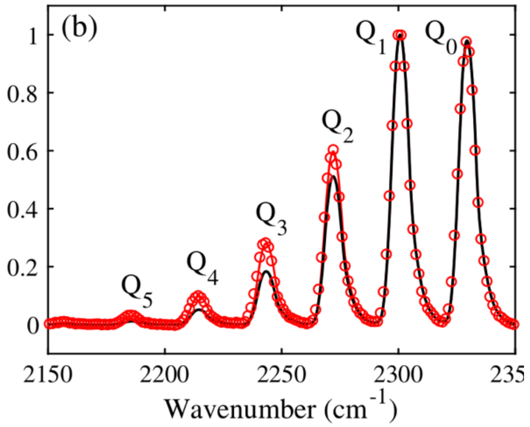




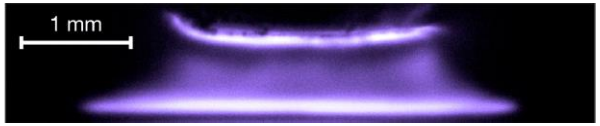
- → Quantify energy level populations directly

CARS measurements of nonequilibrium energy distributions:

Dedic et al., *Optica*, 2017
Pealat et al., *J Appl. Phys*., 1981
Cutler et al., *AIAA Journal*, 2015
Montello et al., *J. Phys. D: Appl. Phys.*, 2015

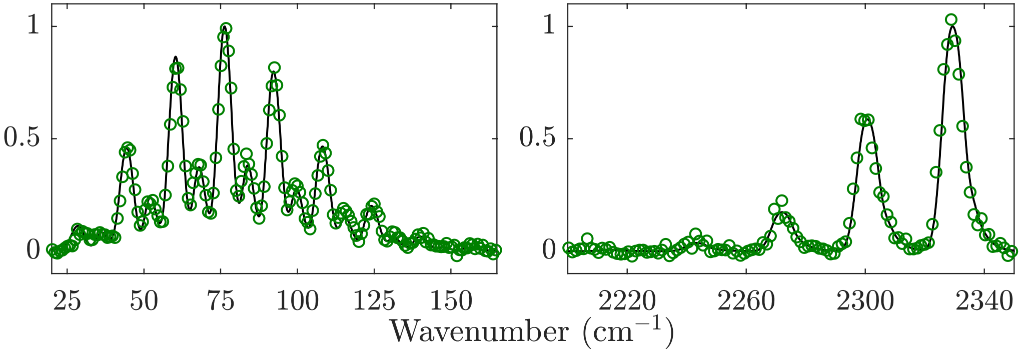


Nonequilibrium energy distributions



13% N_2, 34 Td
RF voltage supply

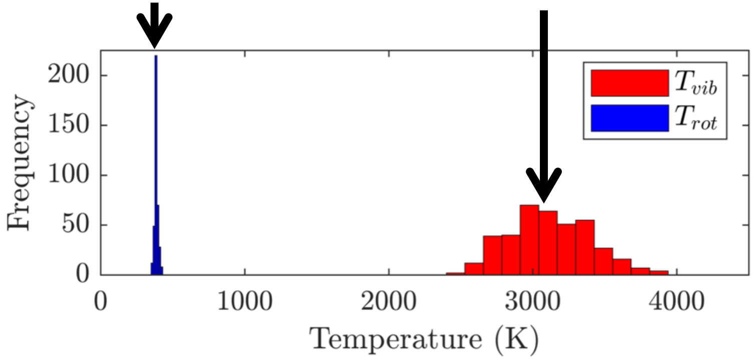
T_{rot} = 390 K, T_{vib} = 3460 K



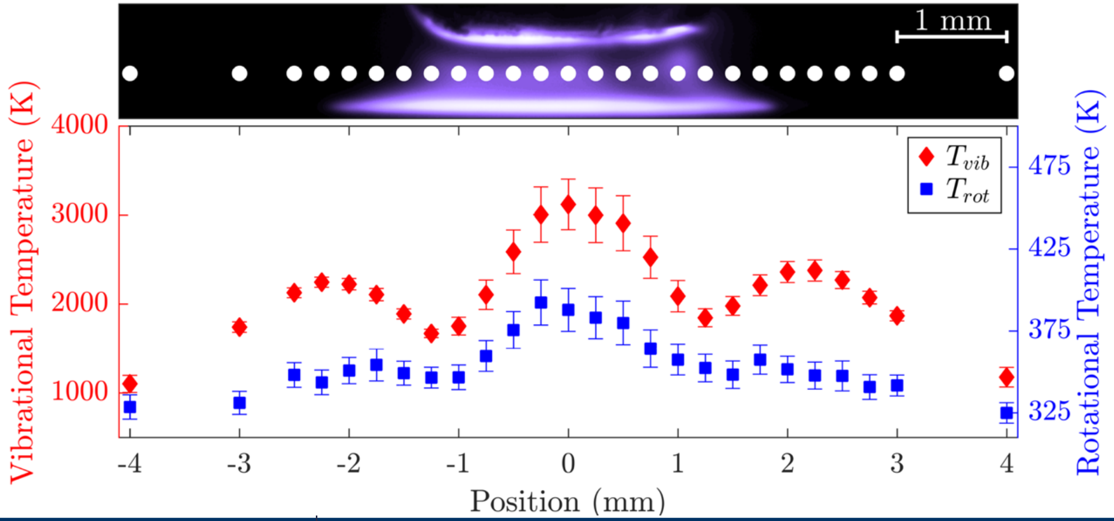
400 single-shot spectra

T_{rot} = 388 K

T_{vib} = 3120 K



→ Secondary temperature peak at 2.25 mm likely due to secondary vibrational excitation:

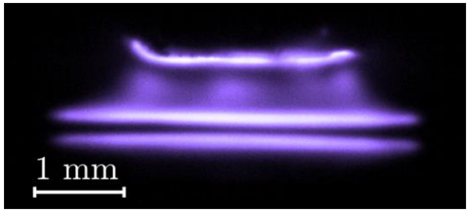


$$N_2(A^3\Sigma_u^+) + N_2(A^3\Sigma_u^+) \rightarrow$$
$$N_2(B^3\Pi_g) + N_2(X^1\Sigma_g^+, v = 8)$$

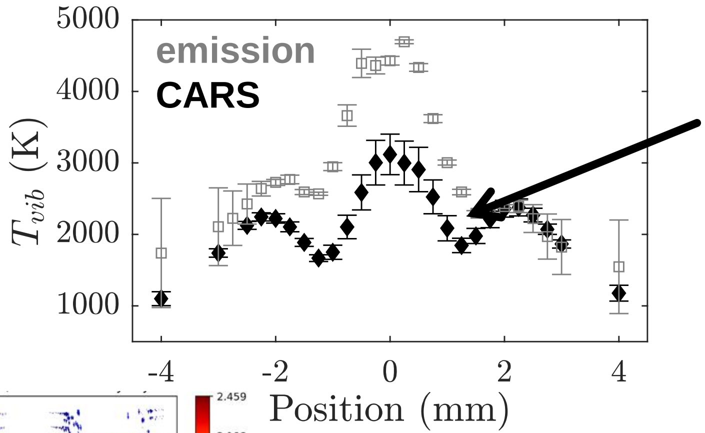
Dedic et al., *Optica*, 2017

Nonequilibrium energy distributions

Compare CARS measurements of $N_2(X)$ to excited electronic state energy distributions to examine secondary rise in temperature post-plasma.

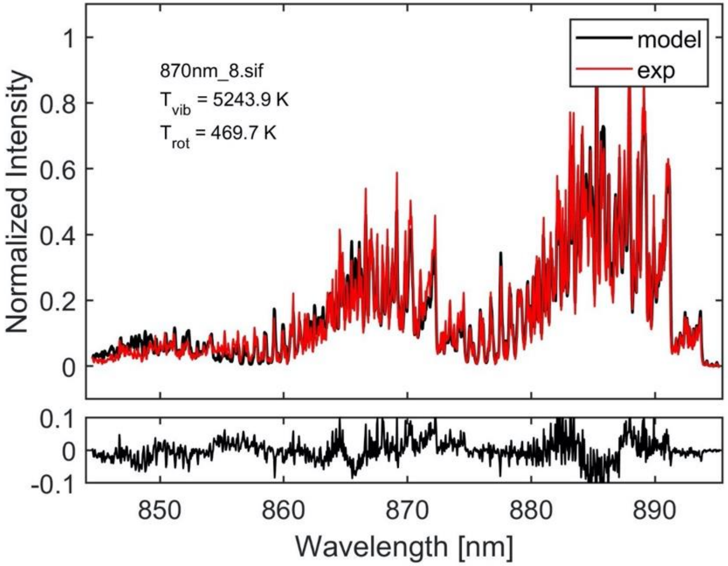


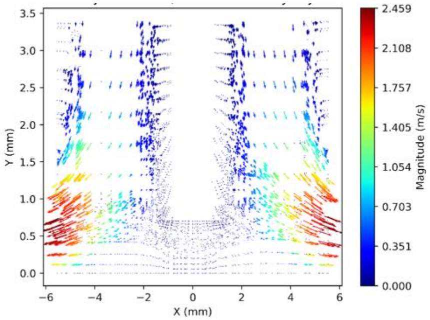
- Plasma emission is path-integrated, but will bias to filaments
- CARS reflects energy distribution from a volume of gas



No emission signal

$N_2(A \leftarrow B)$ emission





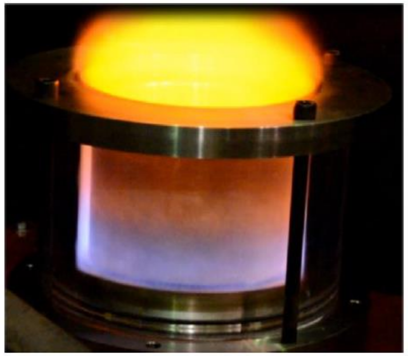
Further kinetic model-based investigation of secondary peak in T_{vib}

Thompson, Michael, Dedic,
In preparation

Transition to a pulsed ns discharge to evaluate fs/ps CARS for accurate measurements in N_2/O_2 and $N_2/CO/CO_2$ mixtures

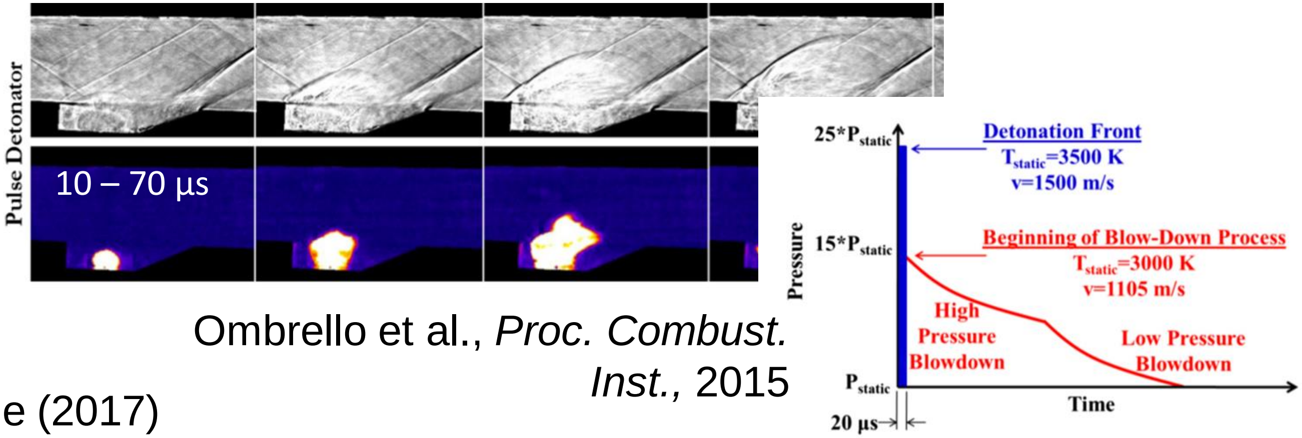
Energy distributions of a decaying detonation

Propulsion Systems



Rankin et al., Combust. Flame (2017)

Scramjet cavity ignition



Ombrello et al., *Proc. Combust. Inst.*, 2015

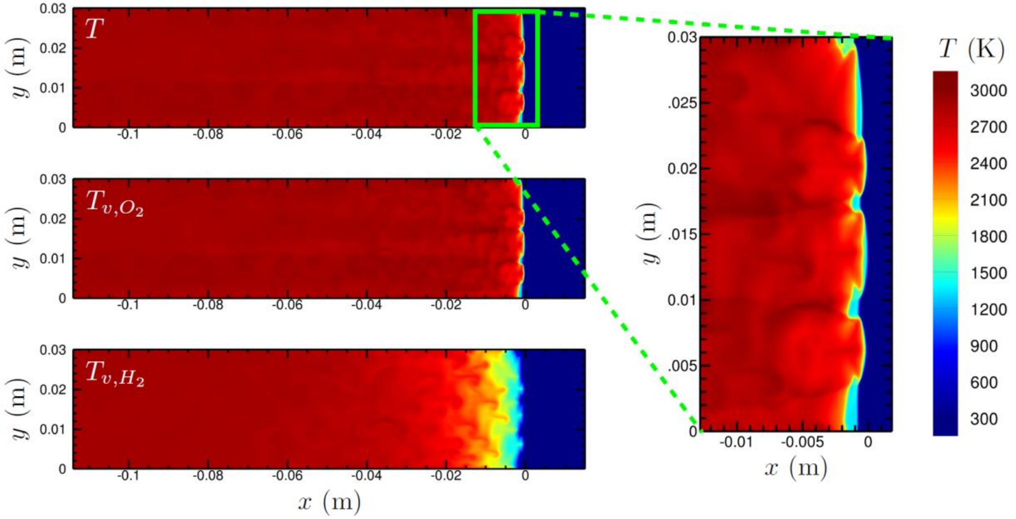
"Extreme environment"

Wide range of…

- T (300 – 4000 K)
- P (1 to 80 bar)

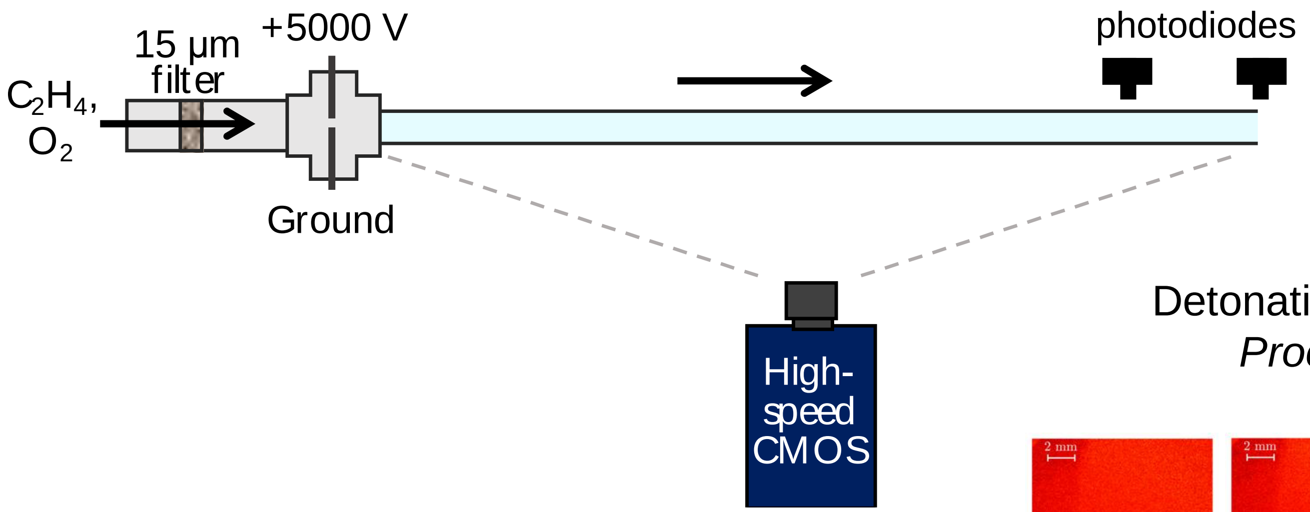
Highly transient process, with multiple relevant time scales:

- μs (wavefront passage, energy transfer) – ms (blowdown)



S. Voelkel et al., *AIP Conference Proceedings*, 2016

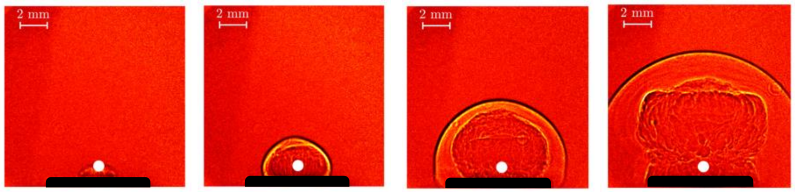
Experimental setup: microscale detonation tube



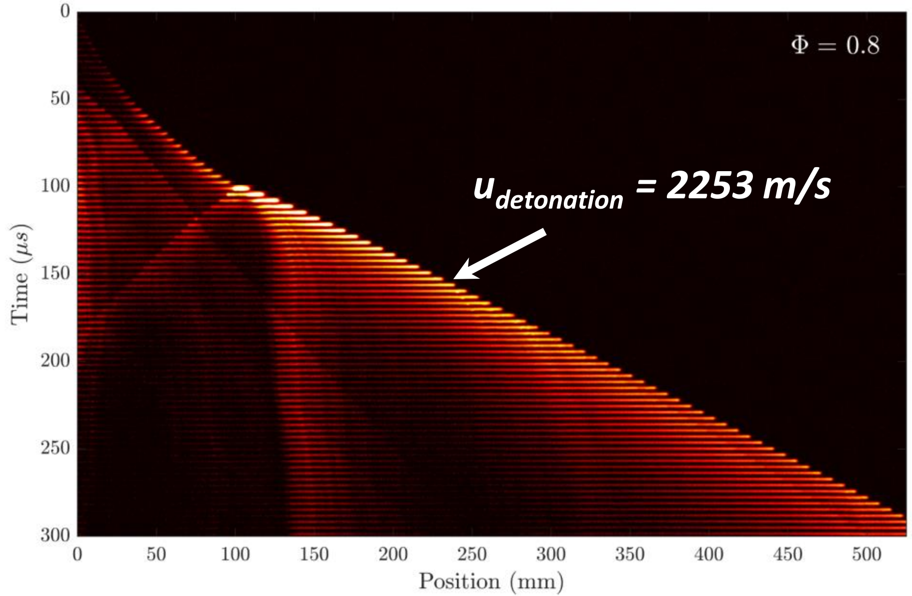
Quartz capillary tube

- L = 0.61 m
- ID = 2 mm
- OD = 8 mm

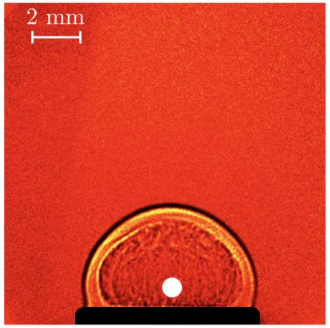
Detonation tube based on Wu et al., *Proc. Combust. Inst.*, 2007

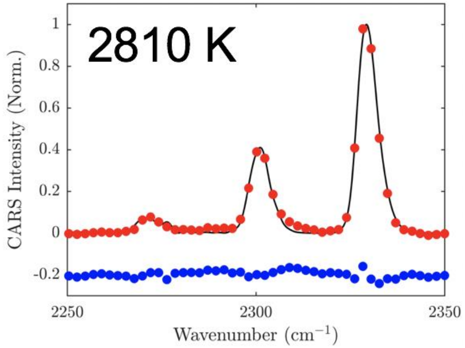


Luminescence within the detonation tube:



CARS measurements at tube exit:

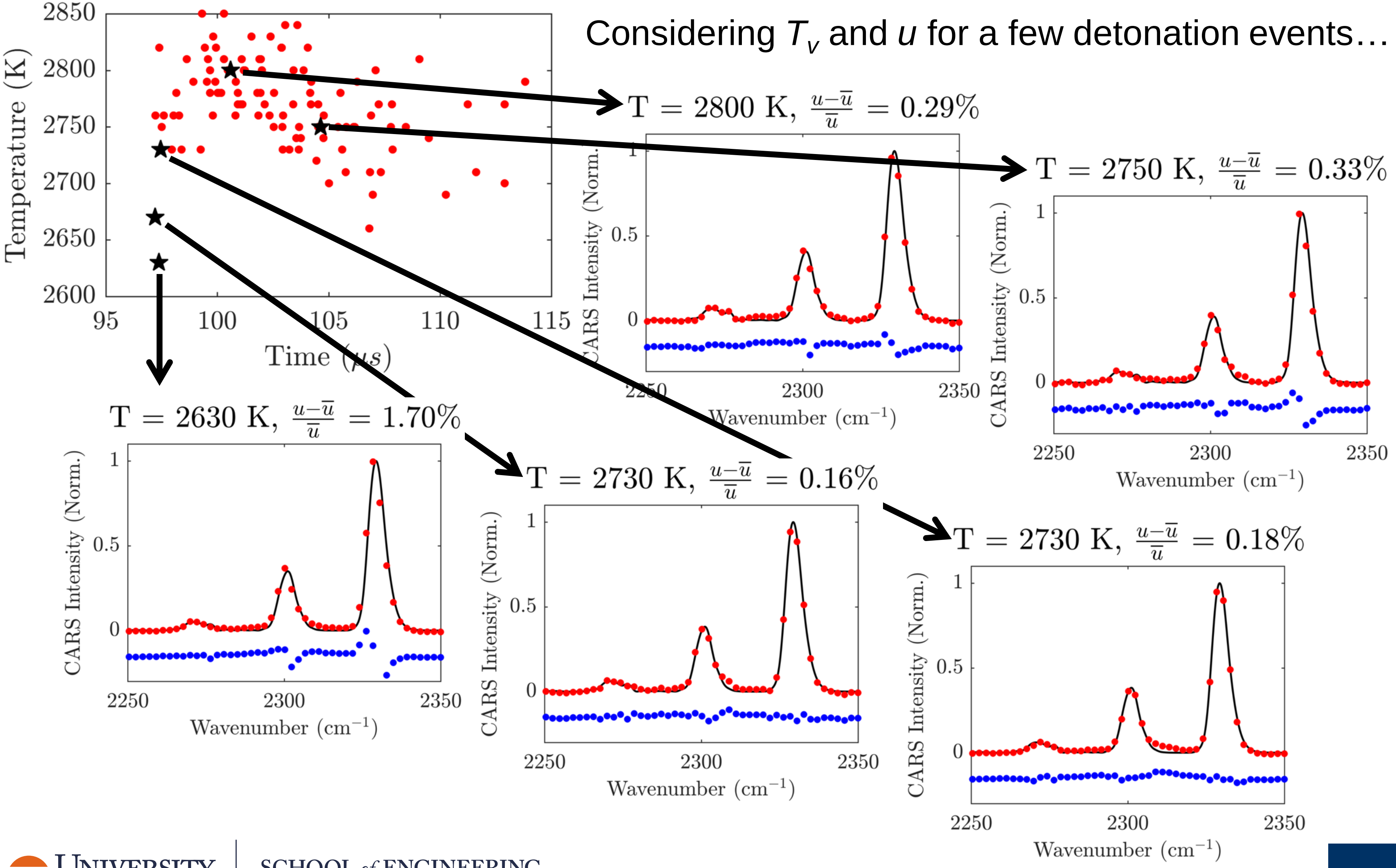




Dedic, Meyer, and Michael, AIAA SciTech 2017
Dedic, Meyer, and Michael, LACSEA 2018

$\phi = 1.0$, 2 slpm N_2

Considering T_v and u for a few detonation events…

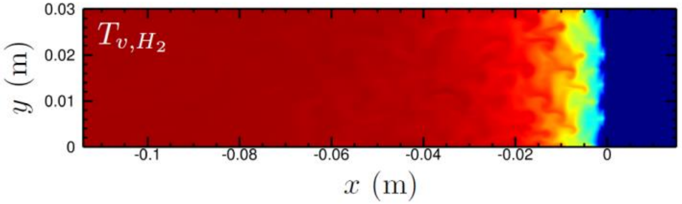


Dedic, Meyer, and Michael, LACSEA 2018

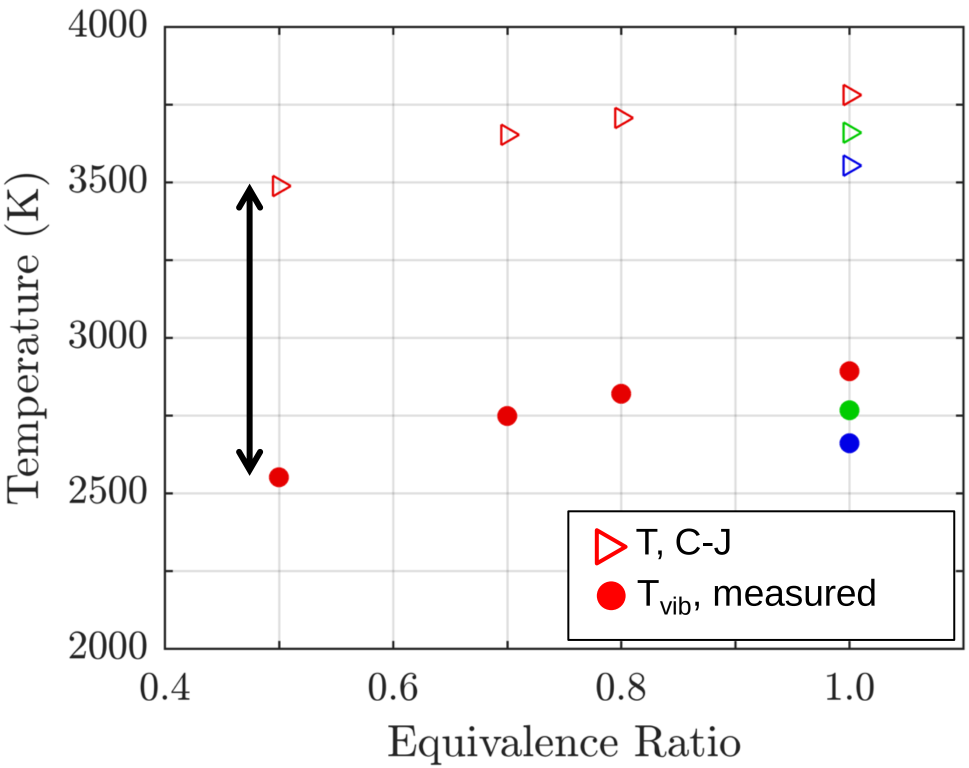
$T_{vib,N2}$ results

- Initial and rapid rise in vibrational T for N_2 with little variation in wavespeed (< 1%)
- Slow decrease in temperature at late times
- Measured vibrational temperature is ~1000 K lower than CJ predictions (heat loss to walls?):

Voelkel et al., 2016

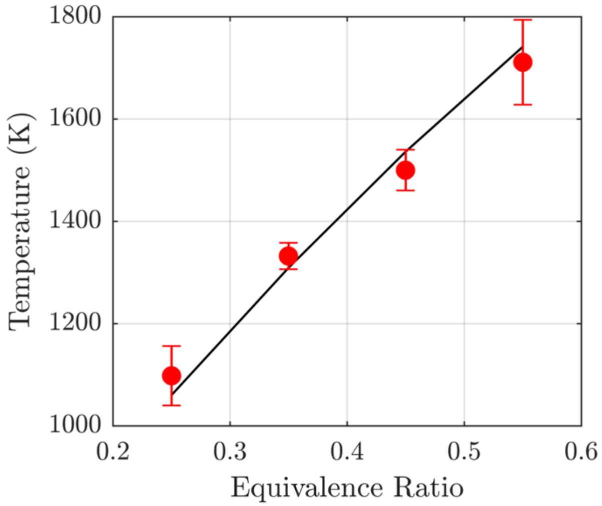


1 mm of predicted nonequilibrium = 5 μs



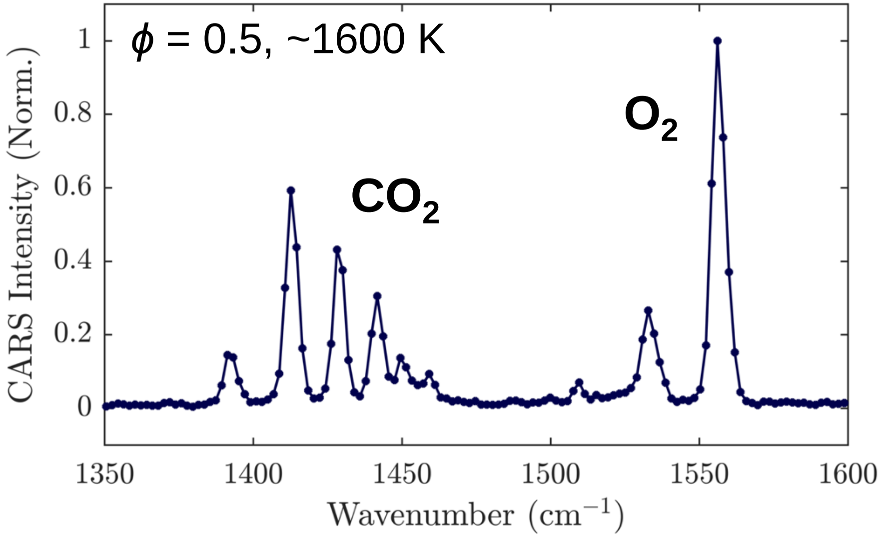
$T_{vib,O2}$ results

Resultant temperatures of an adiabatic flame determined using **O_2 vibrational CARS**:

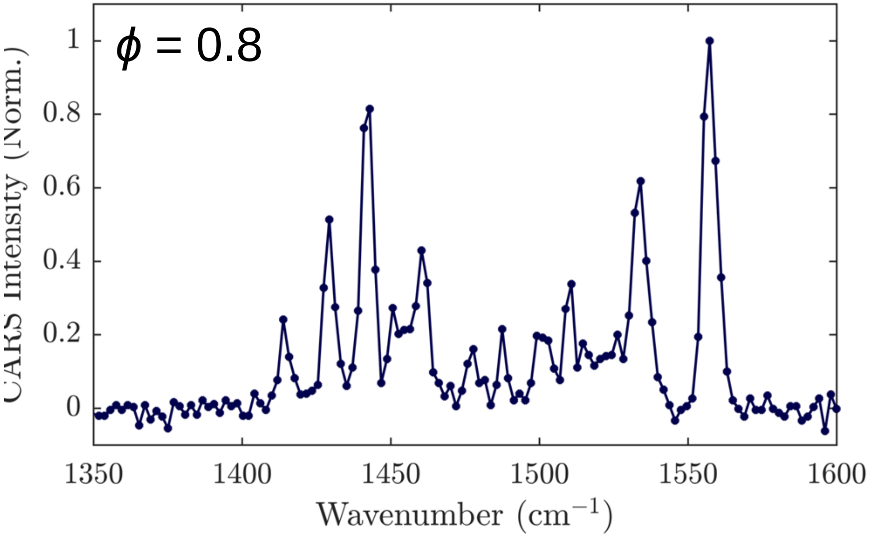


Nearby CARS signal from CO_2 complicates results from detonation blow-down:

Adiabatic flame, C_2H_4/air:

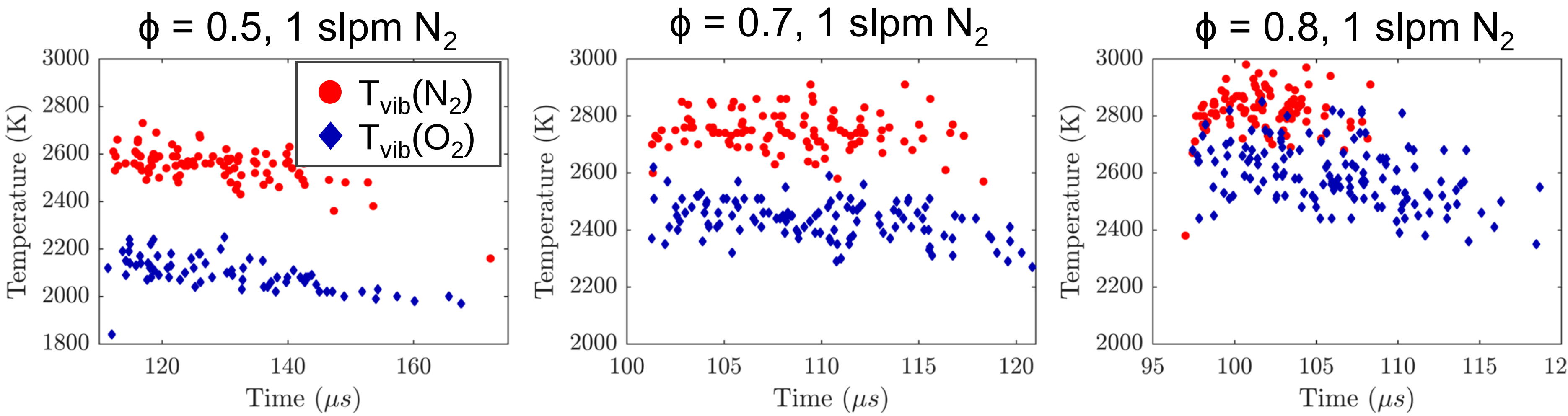


From detonation:



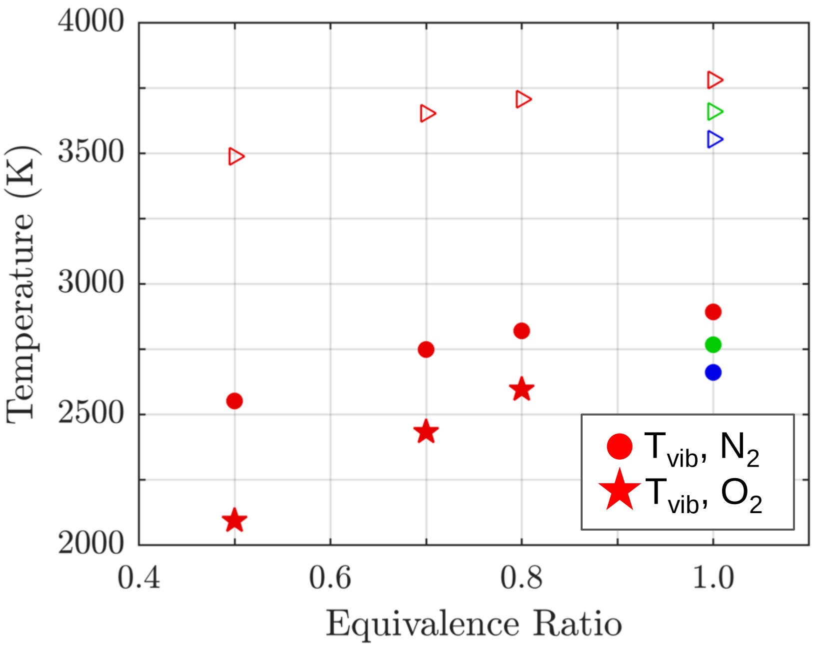
Dedic, Meyer, and Michael, LACSEA 2018

O_2 and N_2 comparison


$\phi = 0.5$, 1 slpm N_2

$\phi = 0.7$, 1 slpm N_2

$\phi = 0.8$, 1 slpm N_2



Lower $T_{vib}(O_2)$ than $T_{vib}(N_2)$, contrary to V-V relaxation timescales:

p (atm)	T (K)	τ_{N2} (μs)	τ_{O2} (μs)
10	2500	6.6	0.21
27	3650	0.83	0.022

Dedic, Meyer, and Michael, LACSEA 2018

Next: Nonequilibrium plasma chemistry

*Validating and optimizing nonequilibrium plasma models will enable improved design of materials and heat shields for Earth/Mars atmospheric entry…**but requires accurate measurements of energy and species in high enthalpy, pulsed ground-based facilities.***

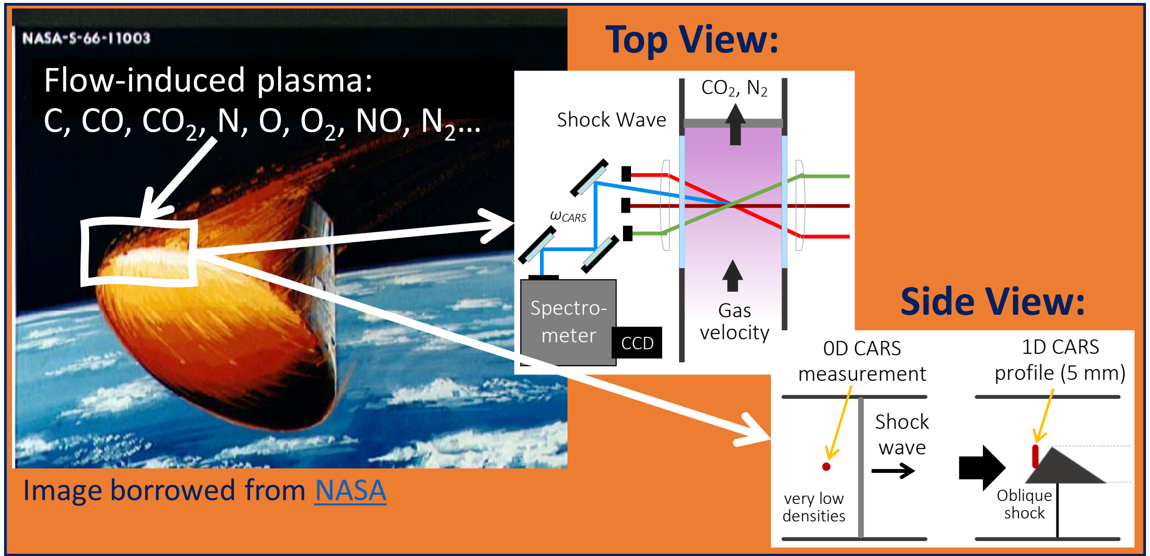


Image borrowed from NASA

Laser system used to measure post-shock wave gases in a shock tube to study key atmospheric entry reactions

Challenges to address:

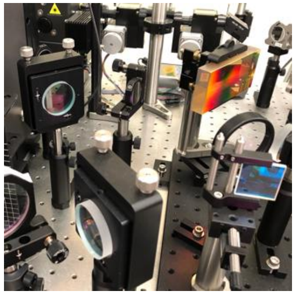
1) Transient flow facilities (NASA EAST)
   - Robust, synchronized system resolving multiple dimensions and species
2) Improve SNR and extract meaningful data from single-shot, low-density environments
3) Accuracy of fs/ps CARS at high-enthalpy, nonequilibrium conditions

NASA Early Career Faculty, 2020

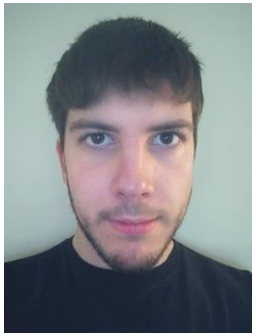
Research Collaborator: Paul Danehy

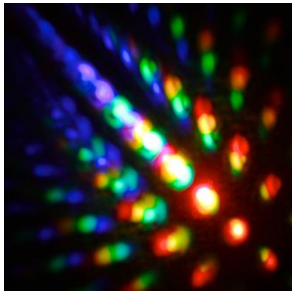
Topic Champions: Aaron Brandis, Mike Barnhardt

University of Virginia Reacting Flow Laboratory



Graduate Students:





Undergraduate Students:

Questions?