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Recent advances in picosecond and hybrid femtosecond/picosecond CARS techniques for gas phase analysis

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Acknowledgement

Funding for this research was provided by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

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Sandia National Laboratories

Albuquerque

Largest US national lab
- > 10,000 people
- ~ 1200 in California
- ~ $2.5B budget

Multi-program
- Nuclear security
- Defense systems
- Energy
- Homeland security

Livermore

WIPP,
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Amarillo, Texas

Tonopah Test Range,
Nevada

Sandia National Laboratories

Combustion Research Facility

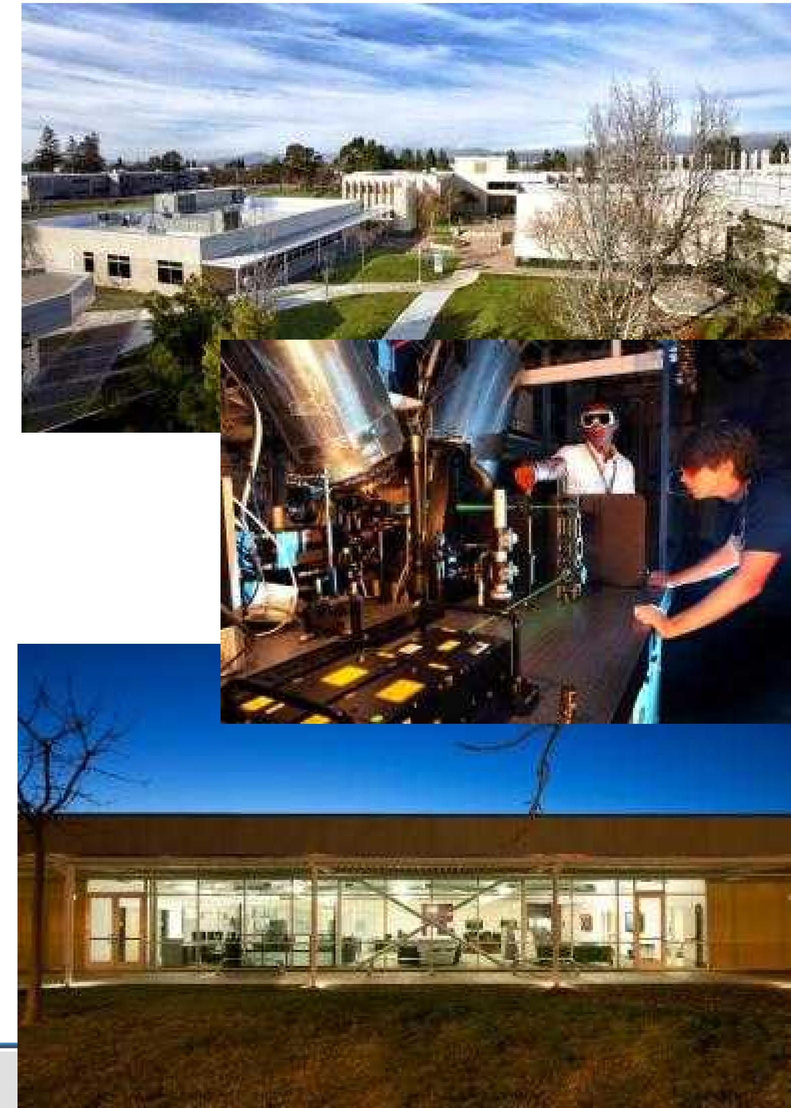
A DOE/BES Collaborative Research Facility dedicated to energy science and technology for the twenty-first century

Key mission

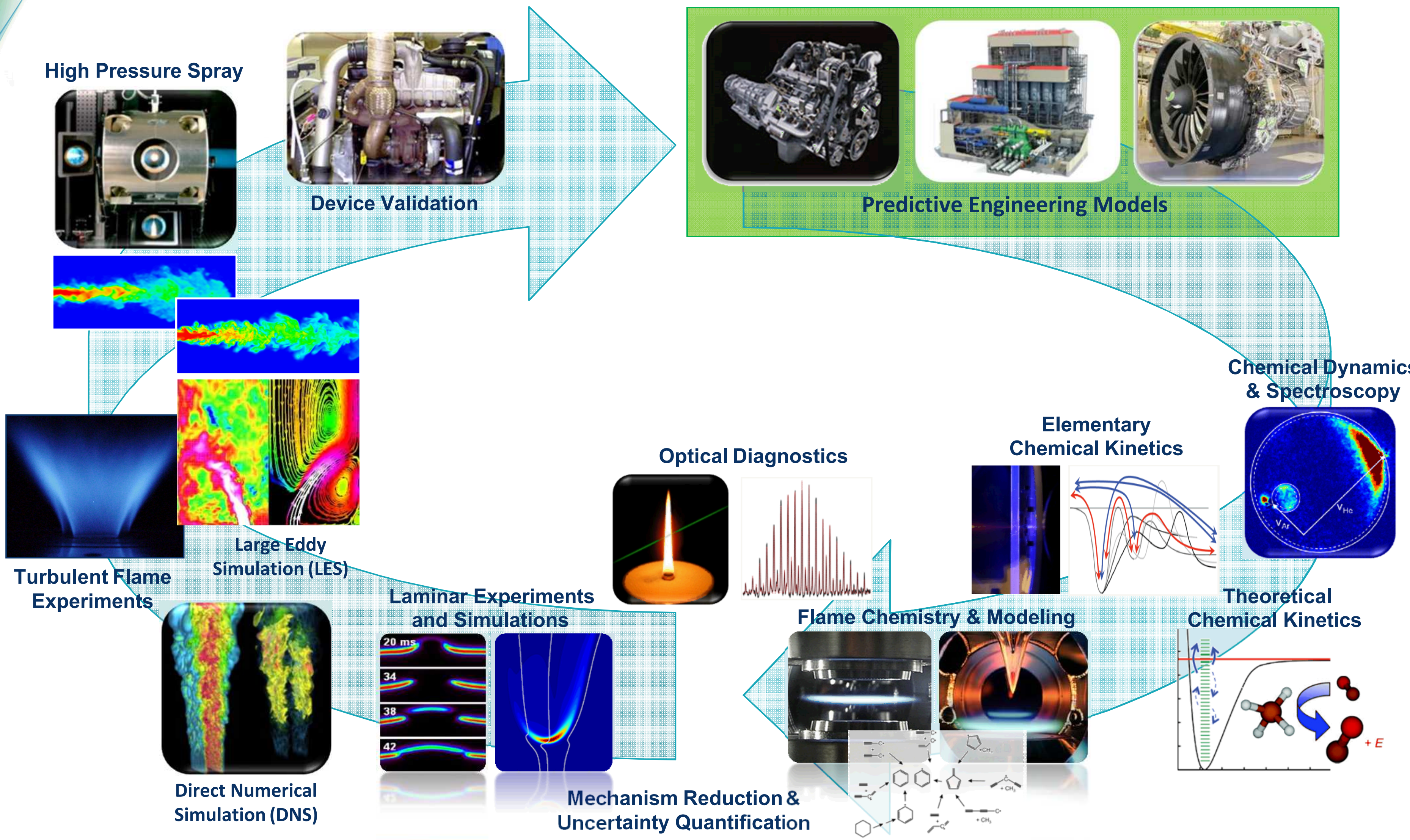
- Provide the science-base needed to develop predictive models for combustion

Facilities

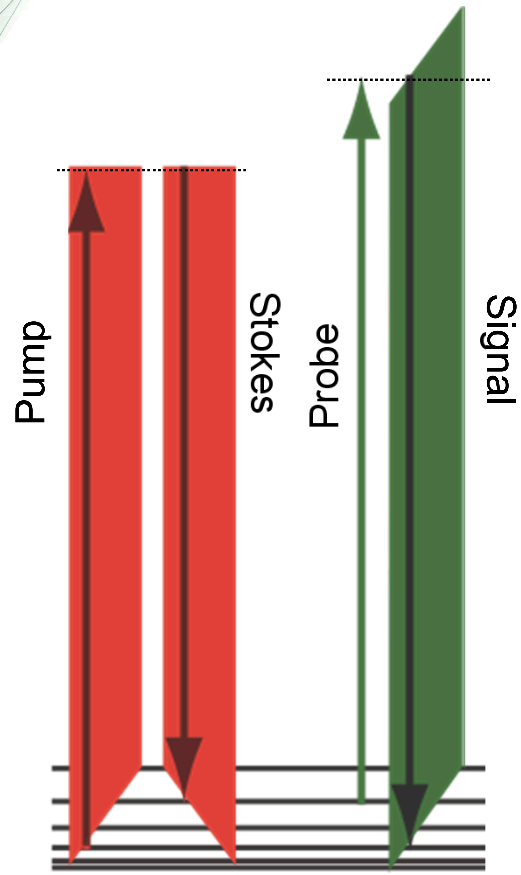
- 82,000-square-foot office and laboratory facility
- 36 highly specialized labs
  - Laser-based diagnostics
  - Combustible and toxic gas handling
  - Computer-controlled safety system
- 8000 square-foot computational laboratory

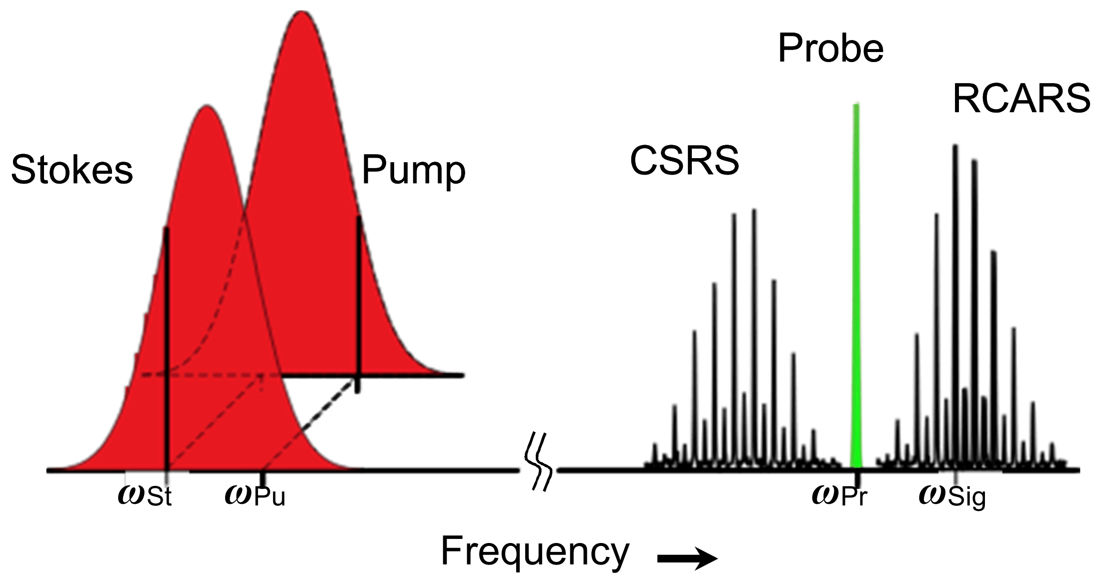


Basic Science Foundation for Predictive Combustion Models



Temperature can be extracted from the measurement of the RCARS spectrum





CARS Signal Depends on:

– Molecular Structure → Chemically Selective
– Geometrical Phase Matching → Spatially Resolved
– Thermal Population Distribution → Temperature Sensitive

Specific Requirement for gas-phase CARS

- Large spatial scale processes require long distance optics
- Low number density requires high pulse energy (100 mJ for ns / 50 µJ for fs)

Combustion analysis

- Turbulent combusting flows typically evolve on a µs time-scale
  - Probe should be pulsed laser (ns/ps/fs) to freeze the dynamics
  - Diagnostic should be capable of single-laser-shot implementation
  - "Video-rate" data requires kHz rate measurement capability

Many implementations for gas phase CARS studies

- Nanosecond CARS
  - Most mature technique
- ★ Picosecond CARS
- Femtosecond CARS
  - Probe-scan CARS
  - CPP fs-CARS
- ★ Hybrid fs / ps CARS
  - Frequency filtering
  - RF-locked ps laser
  - SHBC

A provocation:
Is there a winner for combustion diagnostics?

…we'll finish with the answer

Picosecond CARS

Advantages:

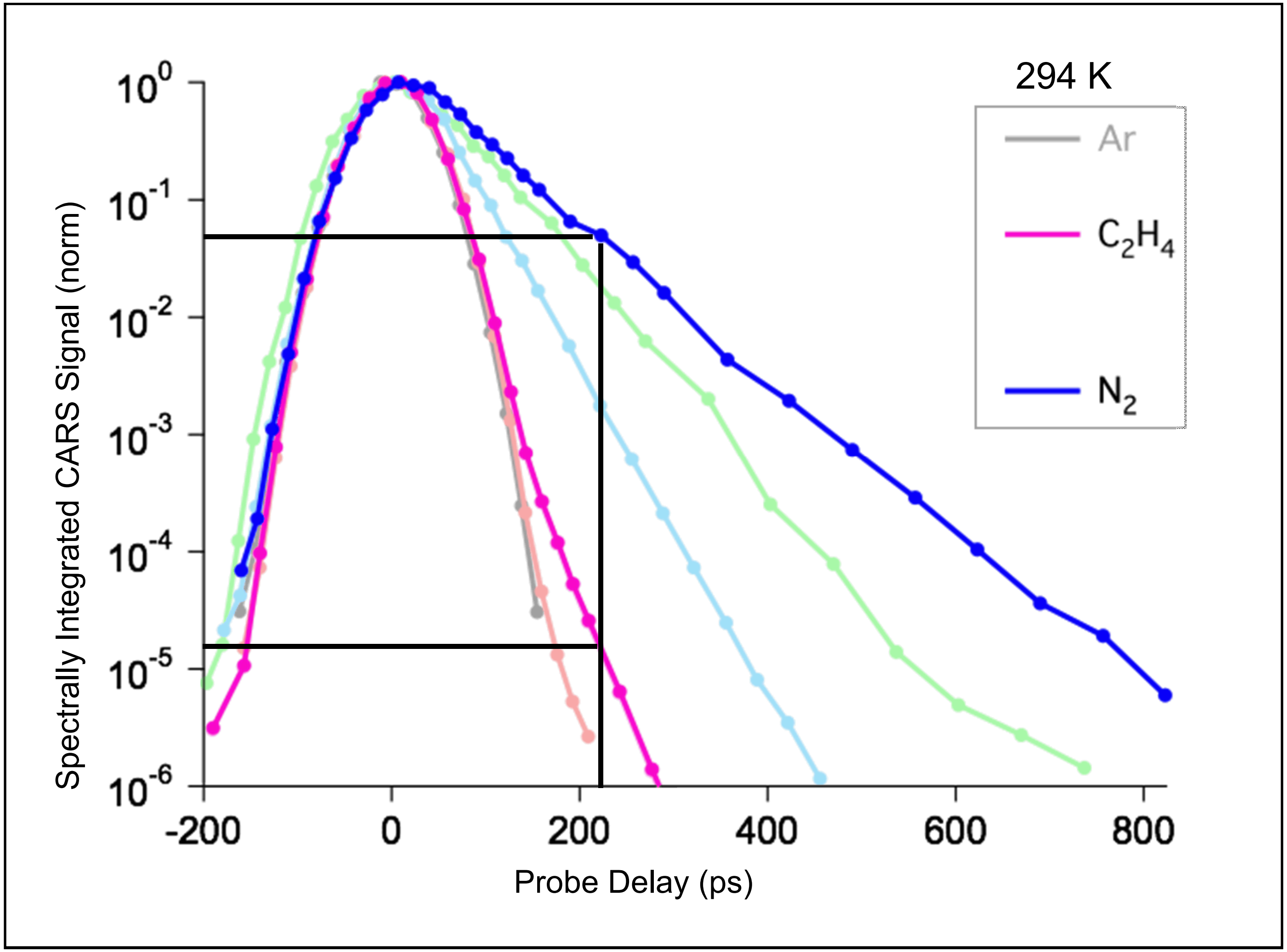
- High peak power with less energy – high signal / low scatter
- Robust and “simple” systems   -field/industrial use
- Direct gas-phase collisional dephasing measurements
- High spectral resolution

kHz rate / 90 ps / 60 mJ

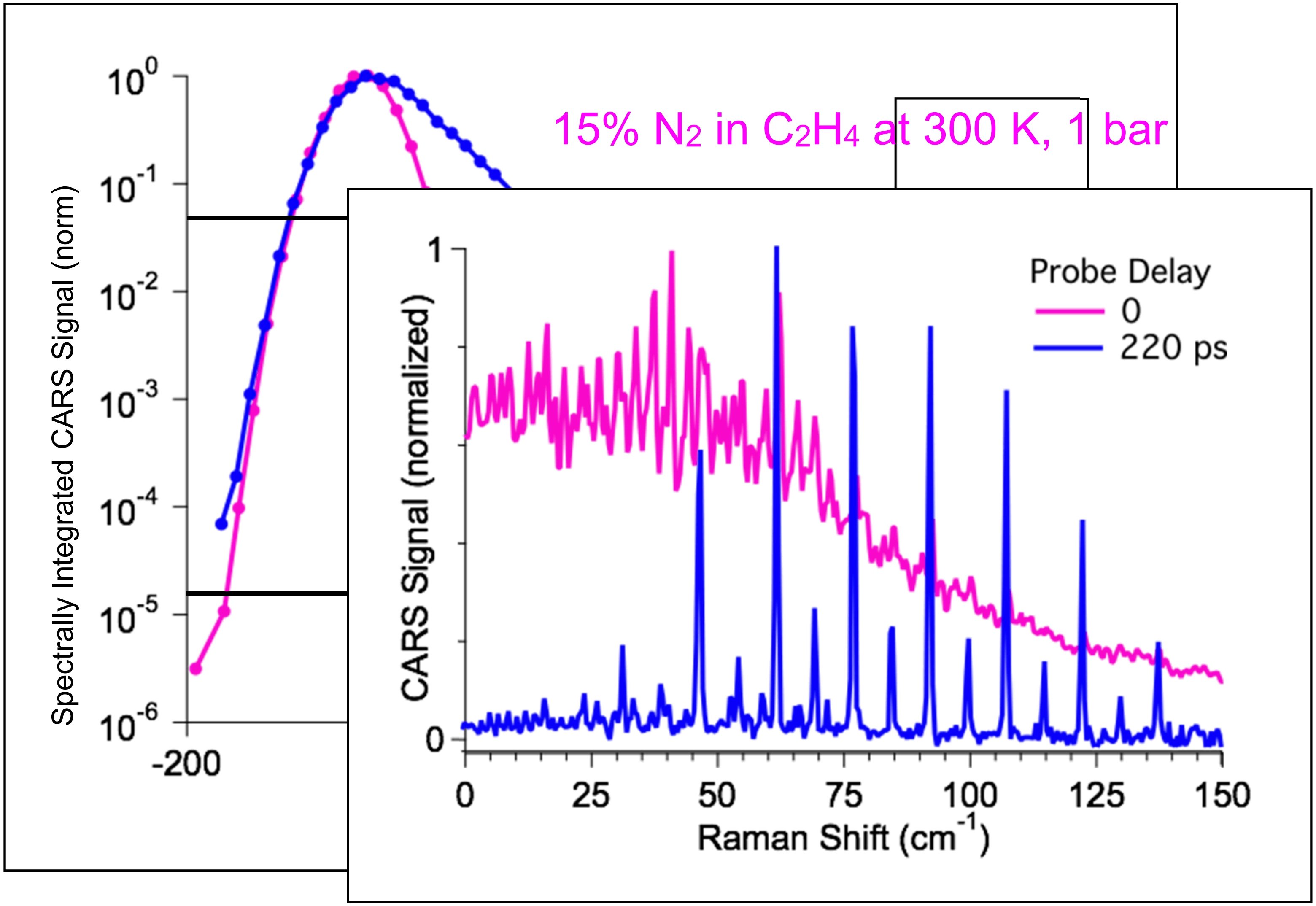
Disadvantages:

- Lower signals than fs approaches
- Sensitivity to collisional environment
- Lower rep rate historically
  - This is rapidly changing

Time-delayed ps-RCARS reveals strong species-dependence for collisional decay rates

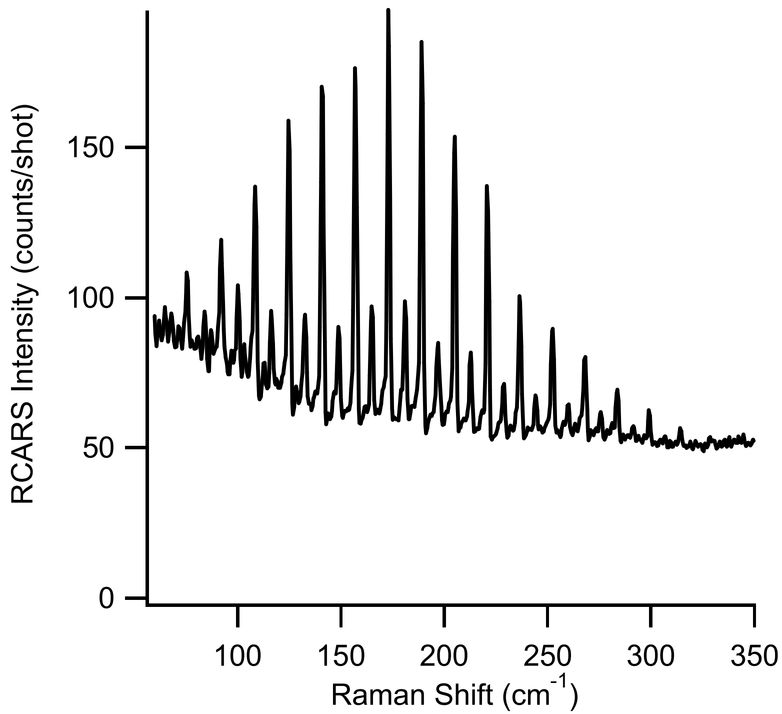


Delayed probe effectively eliminates resonant interference from C_2H_4 in a fuel-rich flow



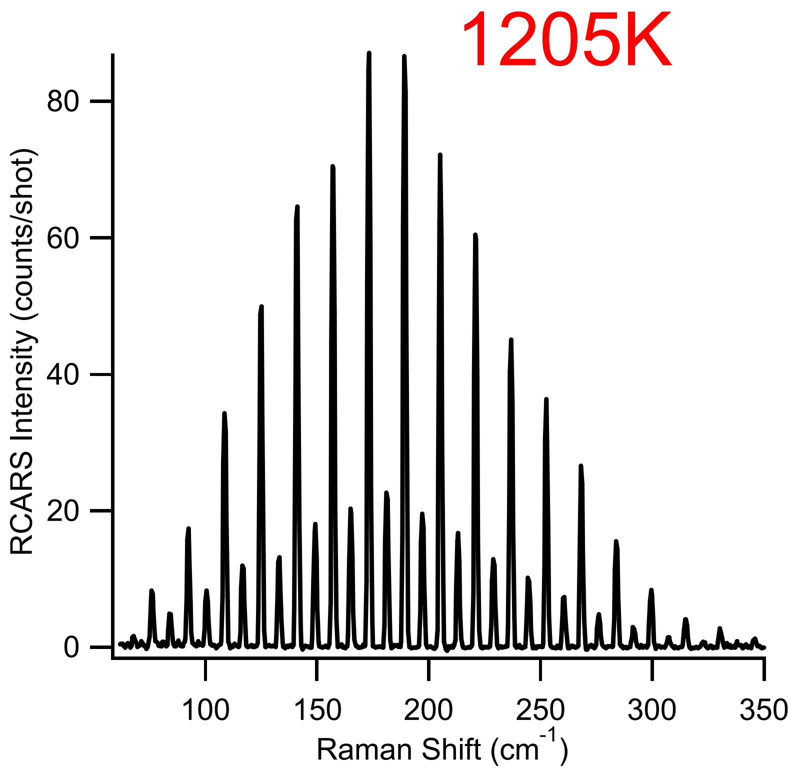
Sooting Ethylene Diffusion Flame

Radial center of ethylene diffusion flame; 0 probe delay

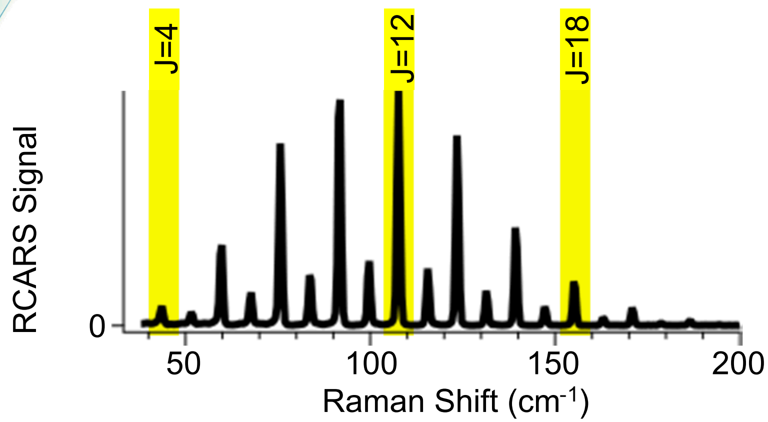


Sooting Ethylene Diffusion Flame

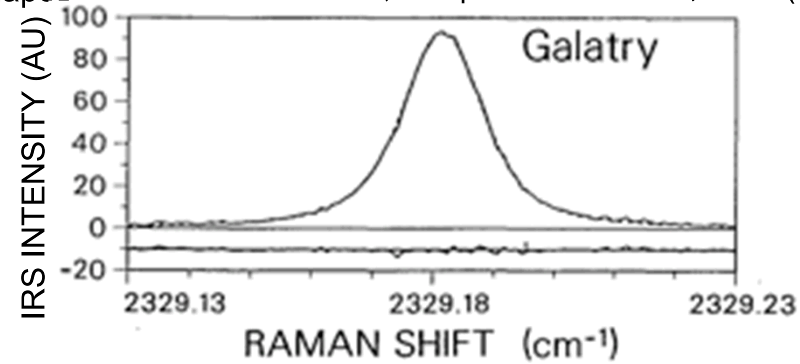
Radial center of ethylene diffusion flame; 150-ps probe delay

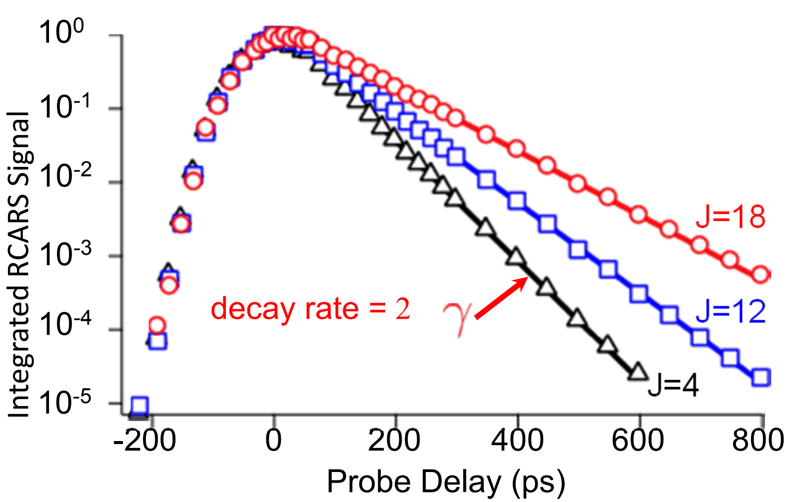


Time-resolved broadband RCARS provides direct multiplexed measurement of pressure-broadening coefficients

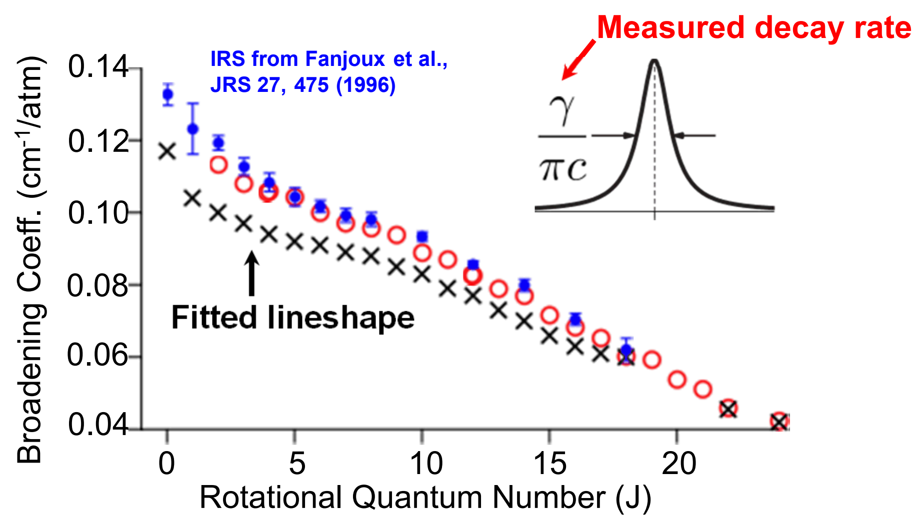


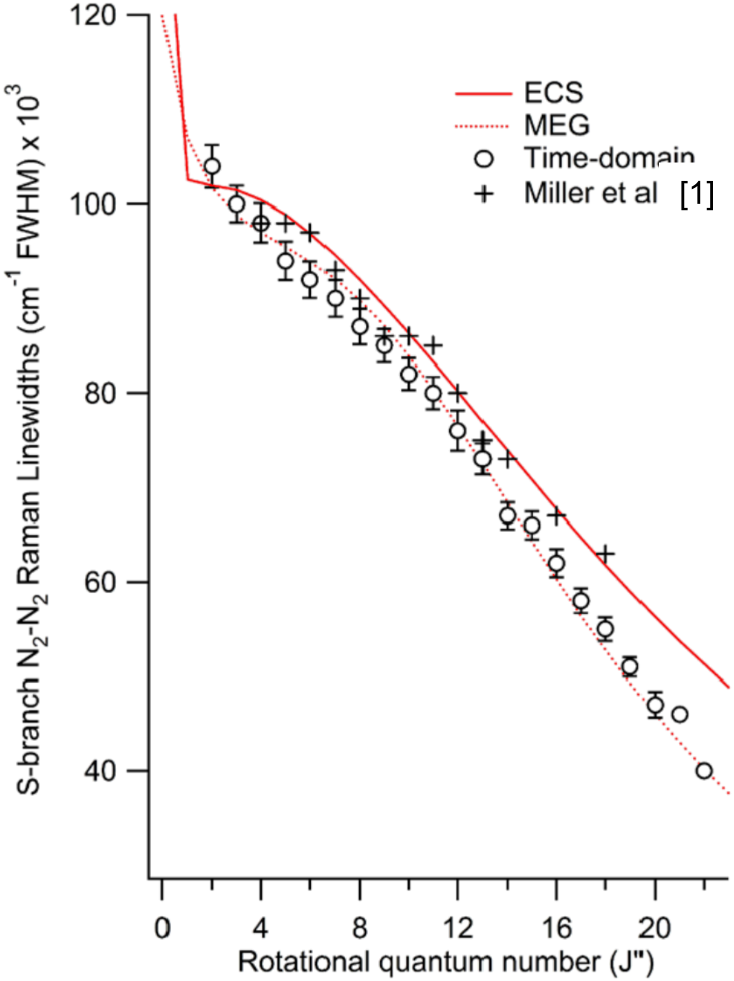
Frequency-domain measurement of J-dependent line shapes Rahn and Palmer, J. Opt. Soc. Am. B 3, 1164 (1986).

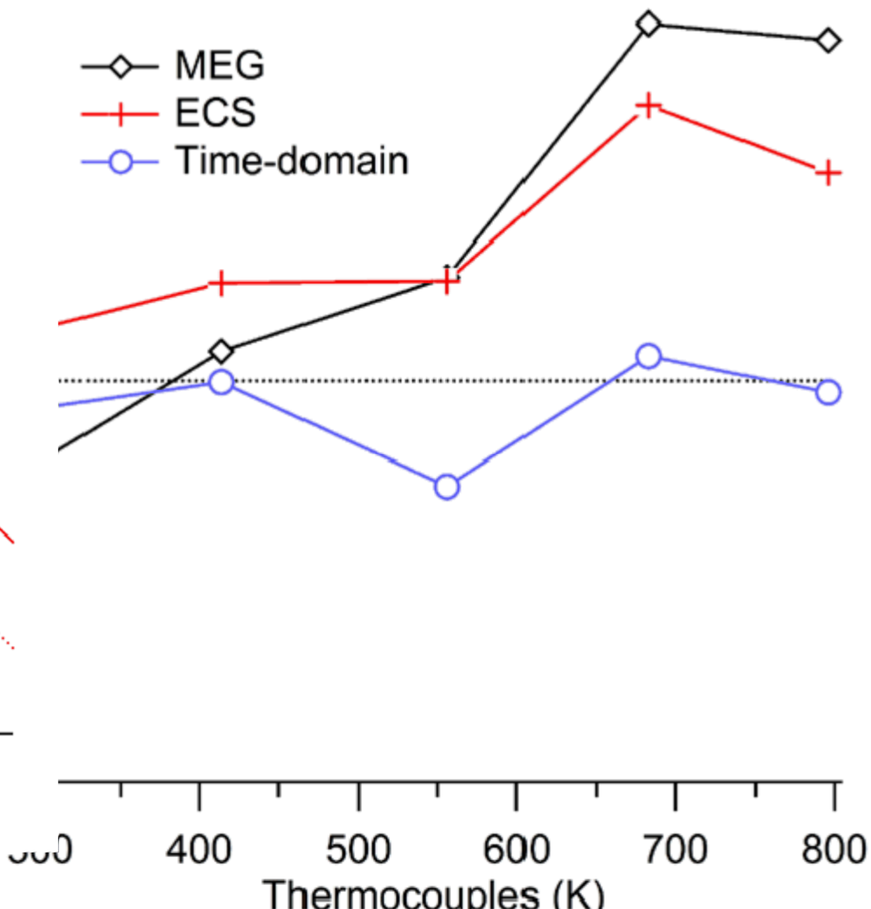




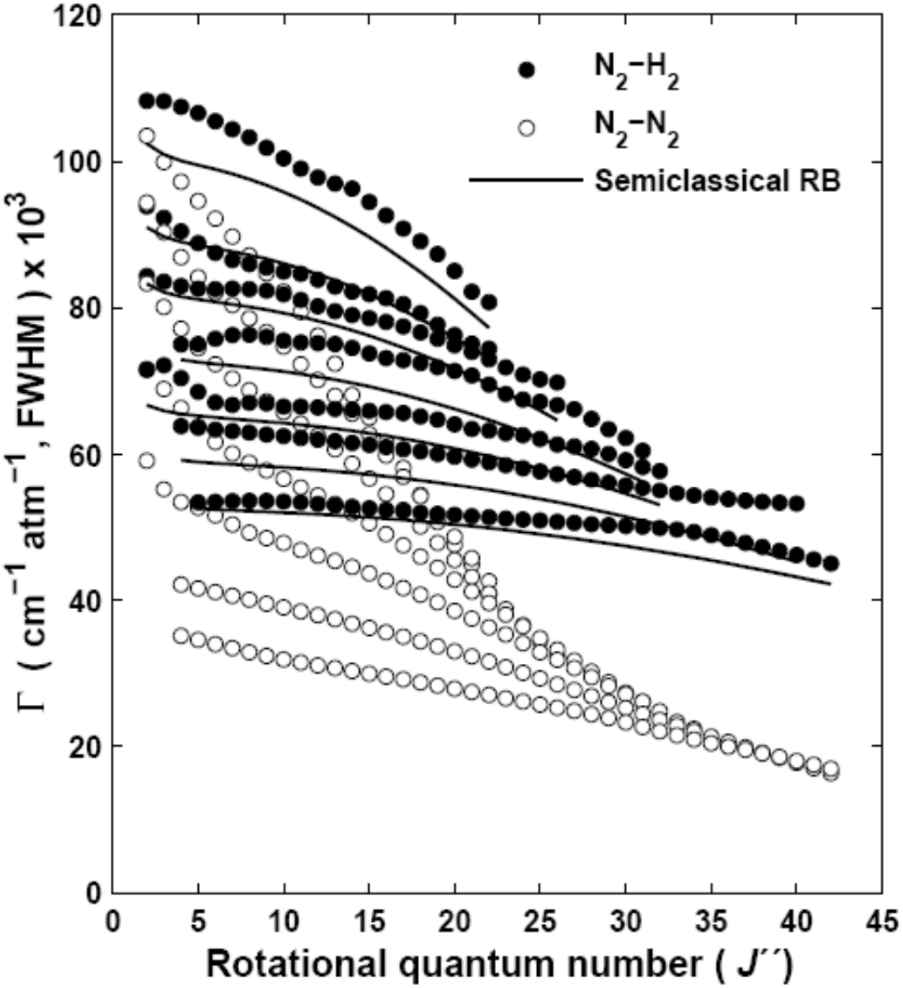
Time-domain measurements of coherence decay rates are multiplexed and only require delay scan.



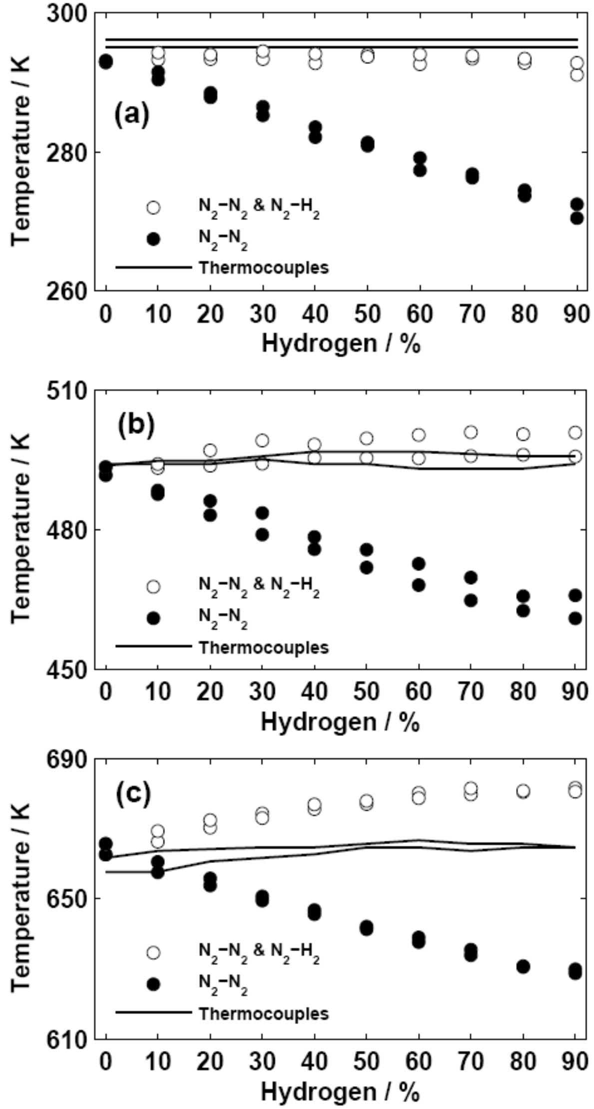




[1] J.D. Miller, S. Roy, J.R. Gord, T.R. Meyer, J. Chem. Phys. **135** (2011)

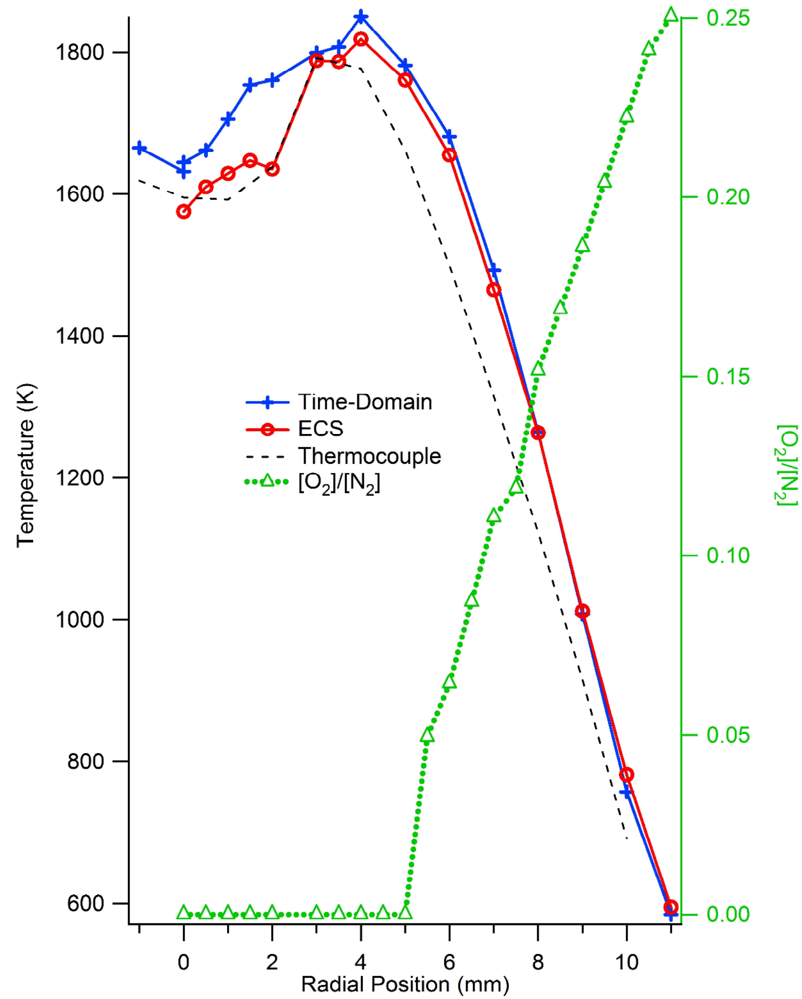


N_2–H_2
N_2–N_2
Semiclassical RB
Γ (cm^{-1} atm^{-1}, FWHM) x 10^3
Rotational quantum number (J´´)

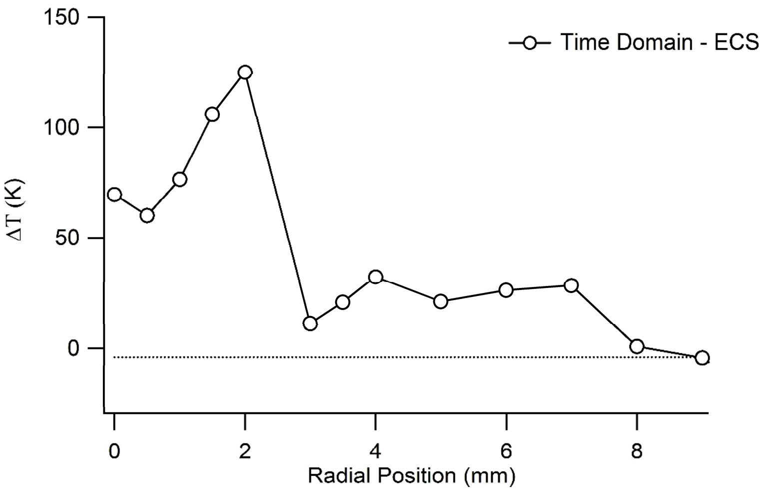


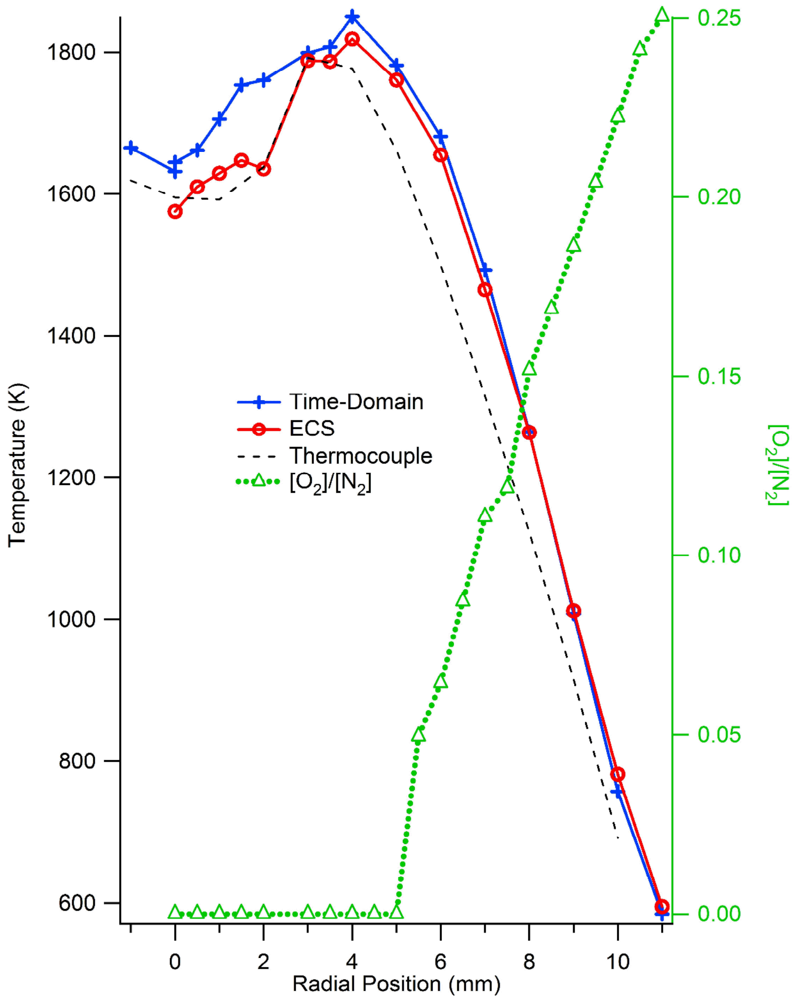
(a)
(b)
(c)
Temperature / K
Hydrogen / %
N_2–N_2 & N_2–H_2
N_2–N_2
Thermocouples

In-situ determination of total broadening coefficents



In-situ determination of total broadening coefficents





In-situ determination of total broadening coefficents

- N_2-N_2 / N_2-O_2
  - C. J. Kliewer, Y. Gao, T. Seeger, J. Kiefer, B. D. Patterson, and T. B. Settersten, Proc. Combust. Inst. **33, 831-838 (2011)**
  - J. D. Miller, S. Roy, J. R. Gord, and T. R. Meyer, J. Chem. Phys. **135 (2011)**
- N_2-H_2
  - A. Bohlin, E. Nordstrom, B. D. Patterson, P.-E. Bengtsson, and C. J. Kliewer, J. Chem. Phys. **137, 074302 (2012).**
- CO_2-CO_2
  - S. Roy, P. S. Hsu, N. Jiang, J. R. Gord, W. D. Kulatilaka, H. U. Stauffer, and J. R. Gord, J. Chem. Phys. **138, 024201 (2013)**
- C_2H_2-N_2 / N_2-C_2H_2 / C_2H_2-C_2H_2
  - P. S. Hsu, H. U. Stauffer, N. Jiang, J. R. Gord, and S. Roy, J. Chem. Phys. **139 (2013)**
- N_2-X In-situ total broadening
  - Y. Gao, A. Bohlin, T. Seeger, P.-E. Bengtsson, and C. J. Kliewer, Proc. Combust. Inst. **34, 3637-3644 (2013)**

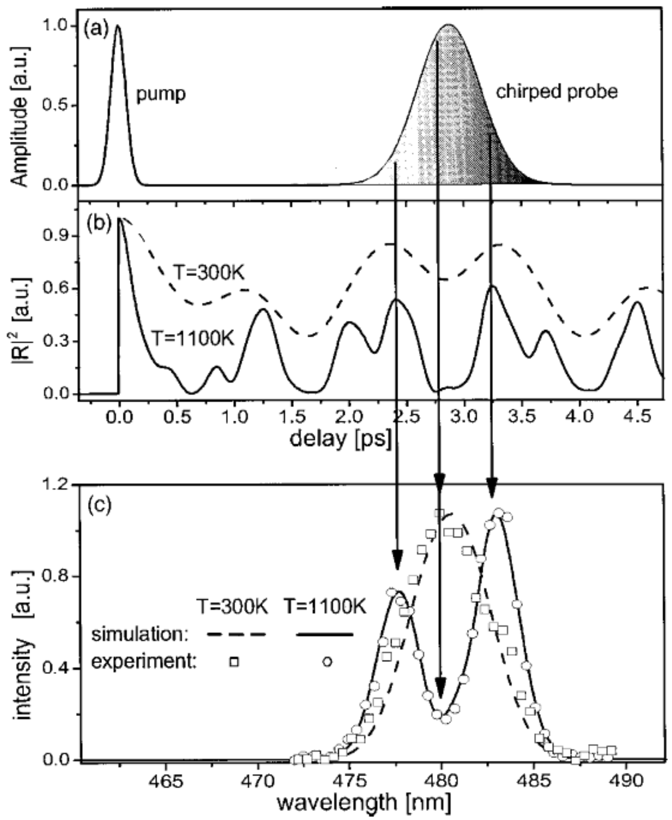
Femtosecond CARS

Advantages:

- Impulsive excitation creates highest possible signal levels
- Collision-independent measurement possible
- Improved shot-to-shot precision
- Shorter integration in the time-domain model (vs. ps-CARS)

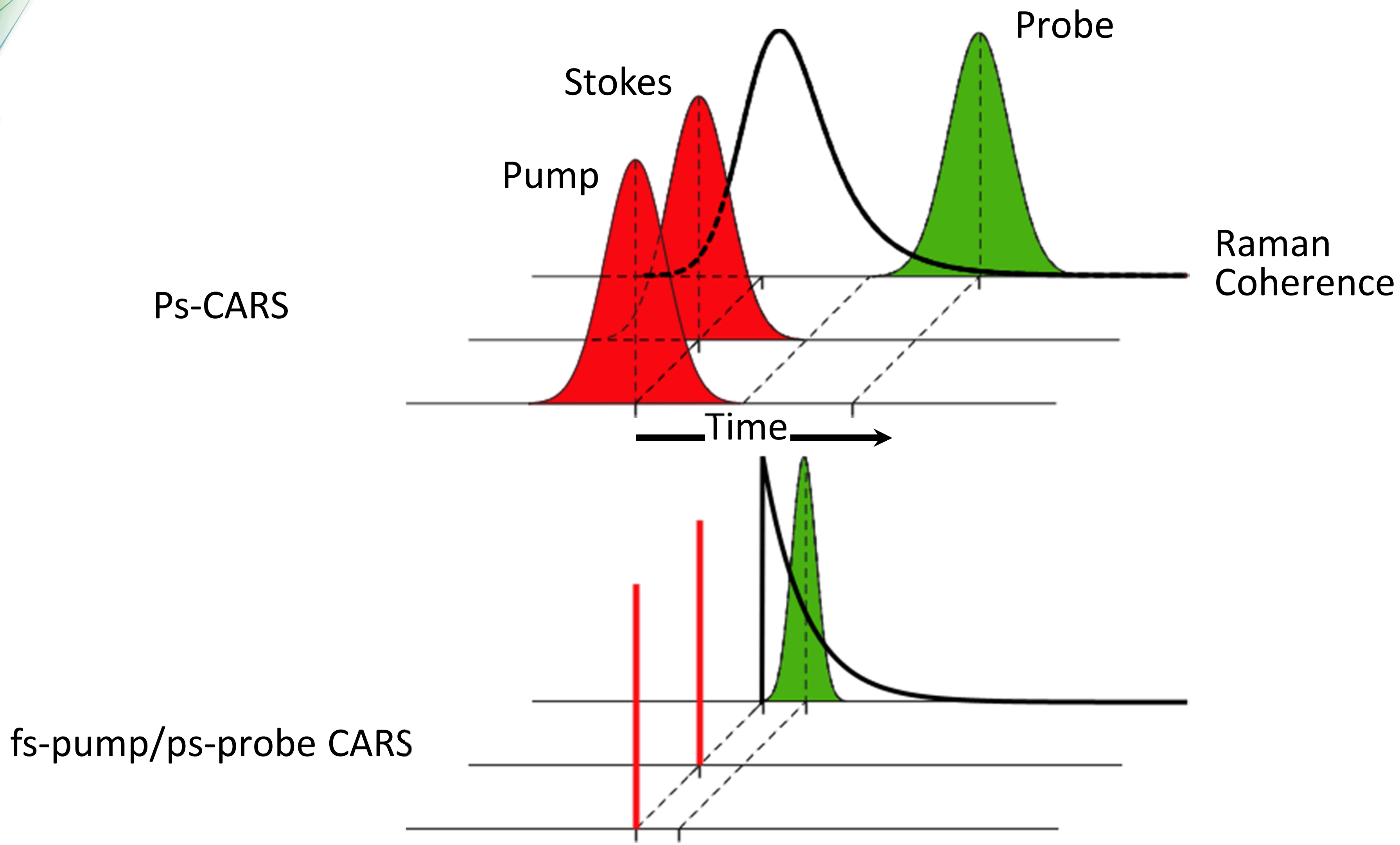
Disadvantages:

- More sensitive/complicated setup than ns- or ps- CARS
- Pulse dispersion concerns / instability
- Sensitivity to probe phase noise?
- Not conducive to 2D imaging



T. Lang and M. Motzkus, J. Opt. Soc. Am. B-Opt. Phys. **19 (2), 340 (2002).**

Femtosecond/Picosecond Hybrid CARS



Femtosecond/Picosecond Hybrid CARS

Advantages:

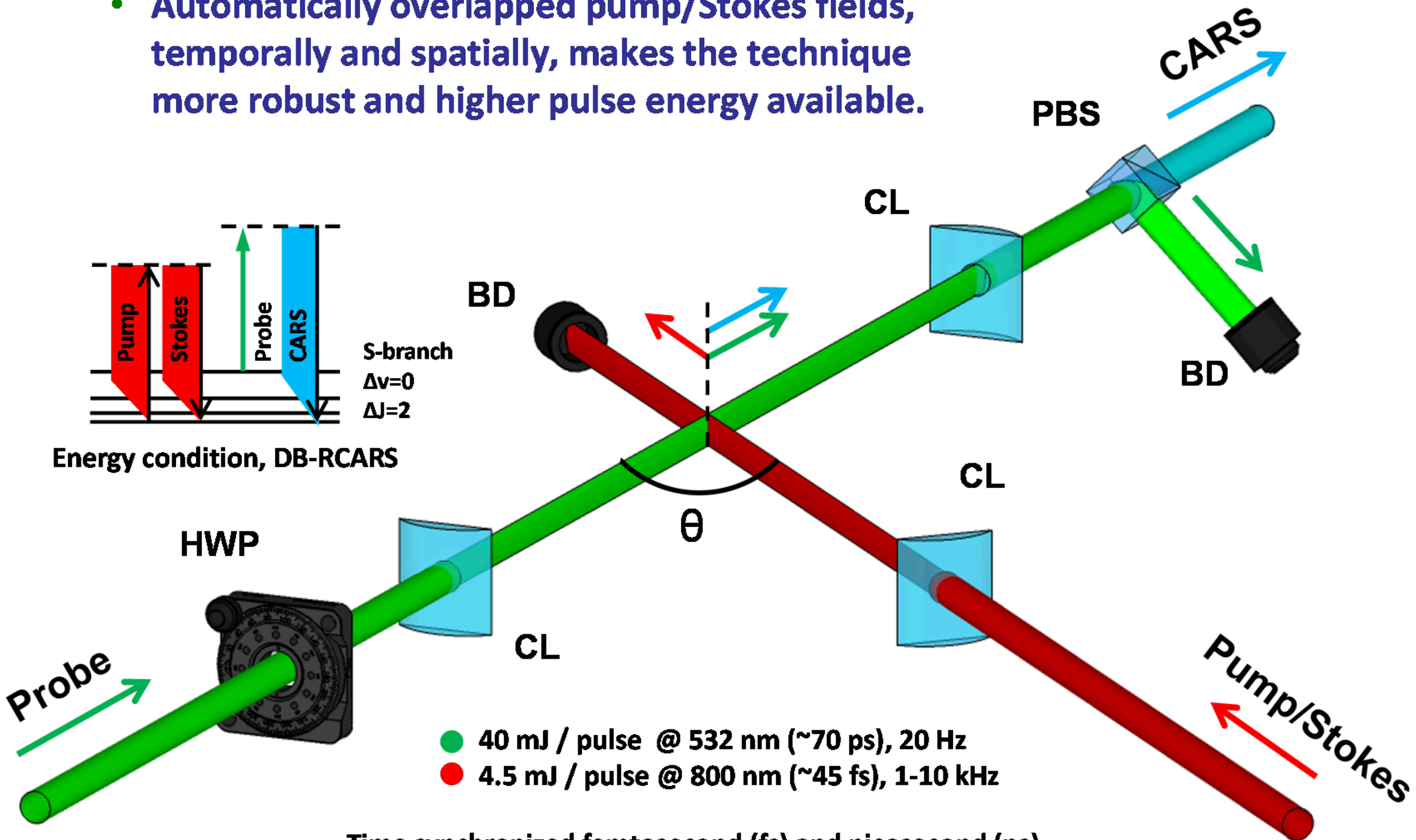
- Impulsive excitation creates highest possible signal levels
- Collision-independent measurement possible (at certain conditions)
- Improved shot-to-shot precision
- High spectral resolution (vs. fs-CARS)
- Shorter integration in the time-domain model (vs. ps-CARS)

Disadvantages:

- More sensitive/complicated setup than ns- or ps- CARS
- Pulse dispersion concerns / instability

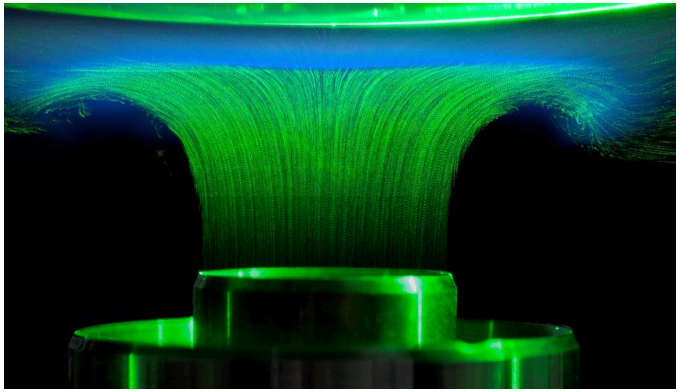
Two-beam hybrid fs/ps 1D-CARS

- Improved spatial resolution (< 50 µm).
- Automatically overlapped pump/Stokes fields, temporally and spatially, makes the technique more robust and higher pulse energy available.



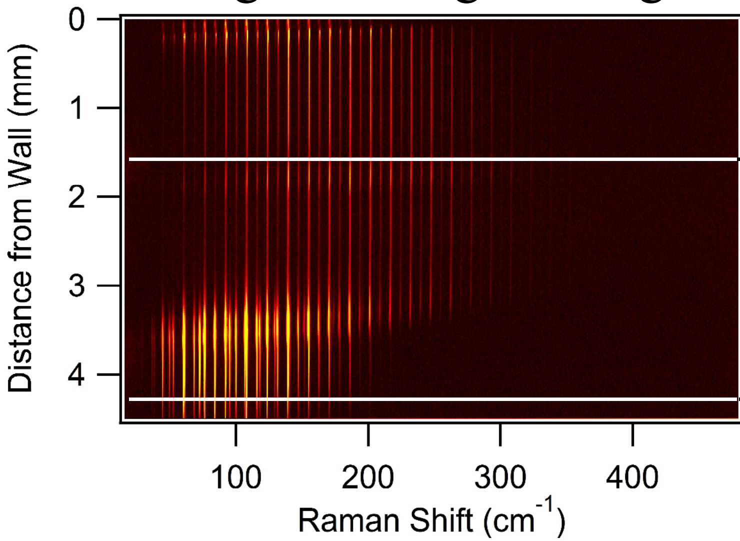
Time synchronized femtosecond (fs) and picosecond (ps) laser system, phase locked to an external 100 MHz RF source

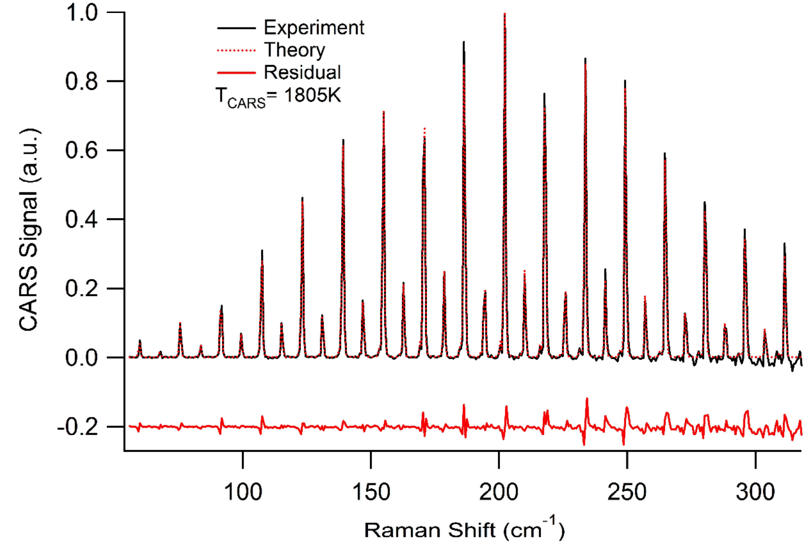
Single-laser-shot 1D-CARS measurements

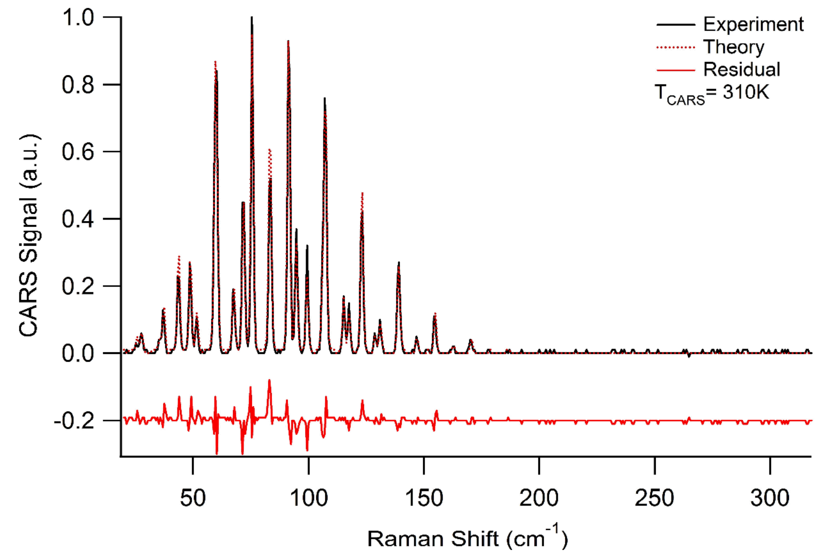


Φ=0.83, *Re*=5000

Single-shot signal image







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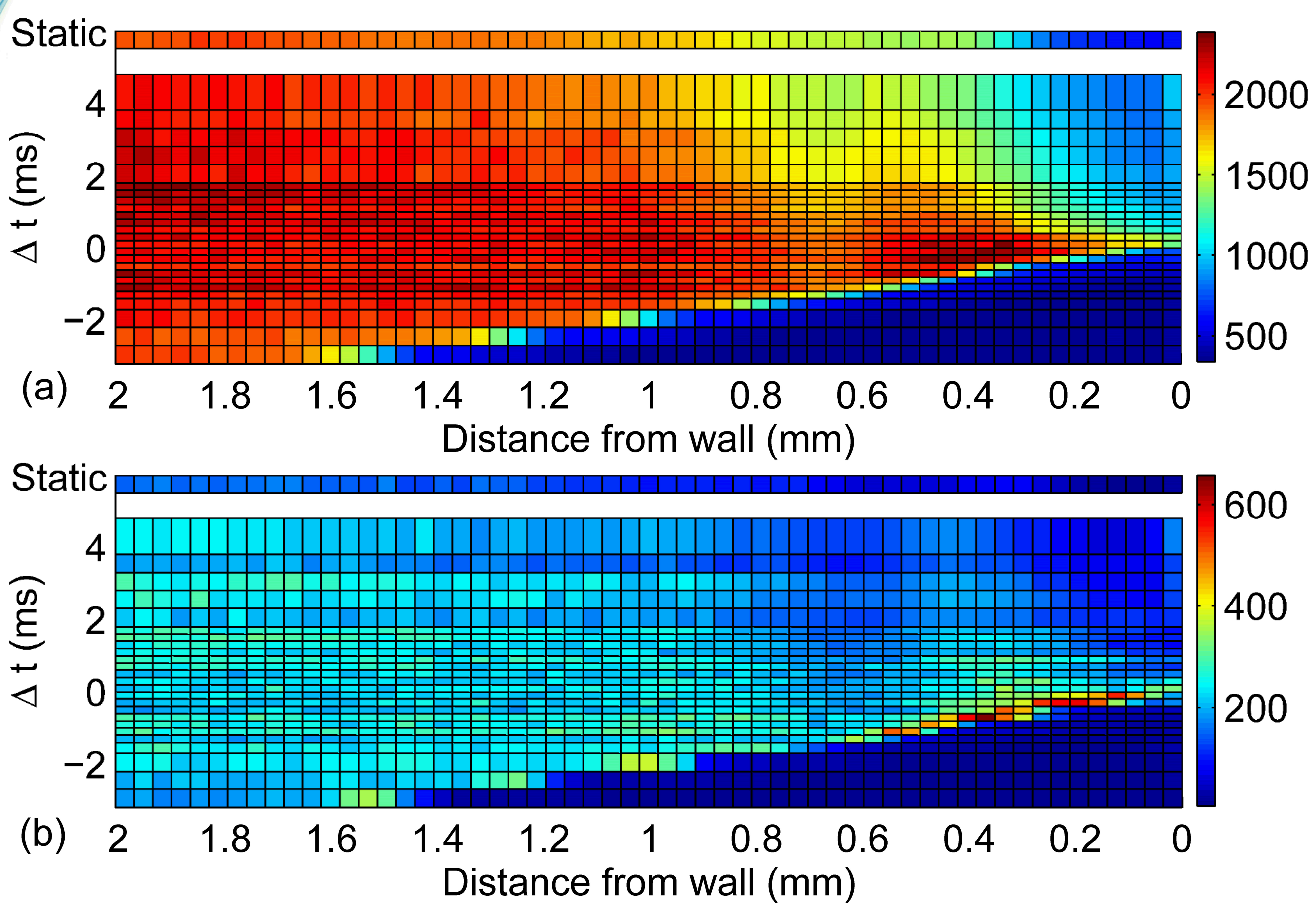
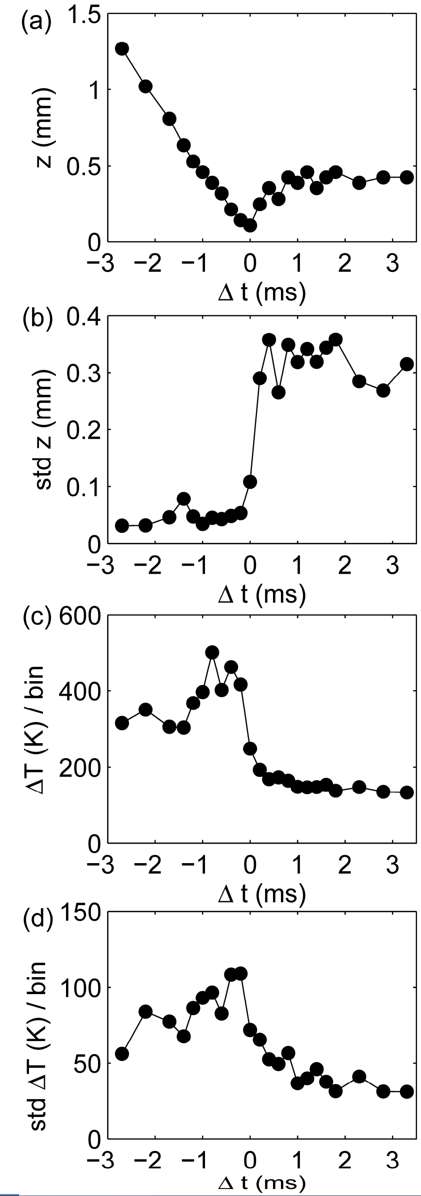


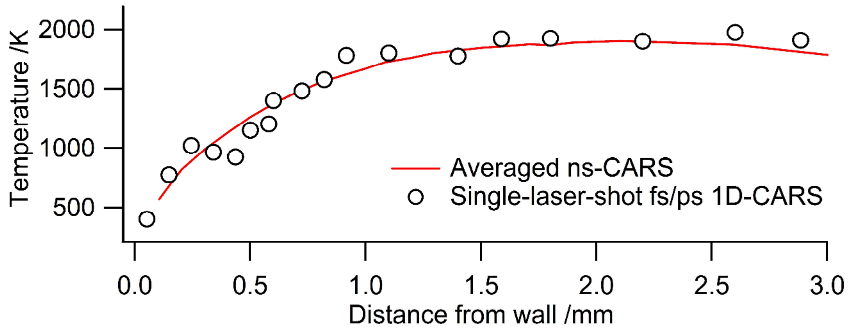


Time- and spatially dependent statistics of the 1D flame front gradient / thickness / position become possible

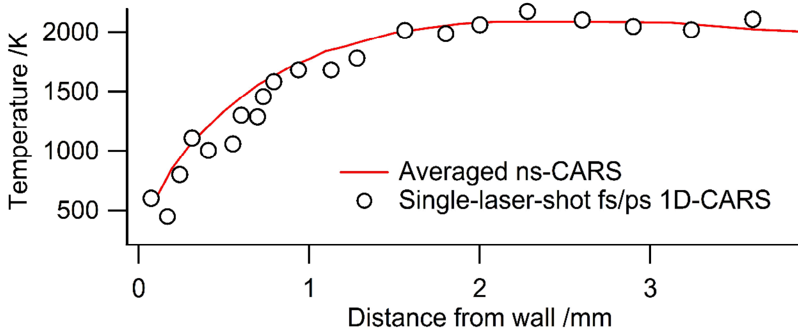
Single-laser-shot 1D-CARS measurements

•Detailed time-resolved studies of transient FWI effects become possible with single-shot thermal field measurements using fs/ps 1D-CARS

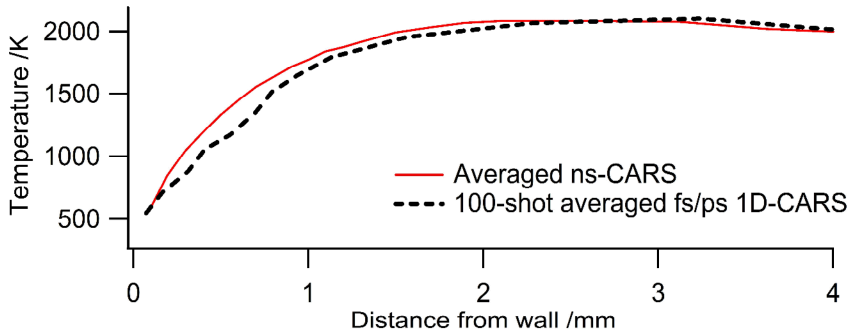
Φ=0.83, *Re* = 5000
Single laser shot



Φ=1.0, *Re* = 5000
Single laser shot



Φ=1.0, *Re* = 5000
100 laser shots



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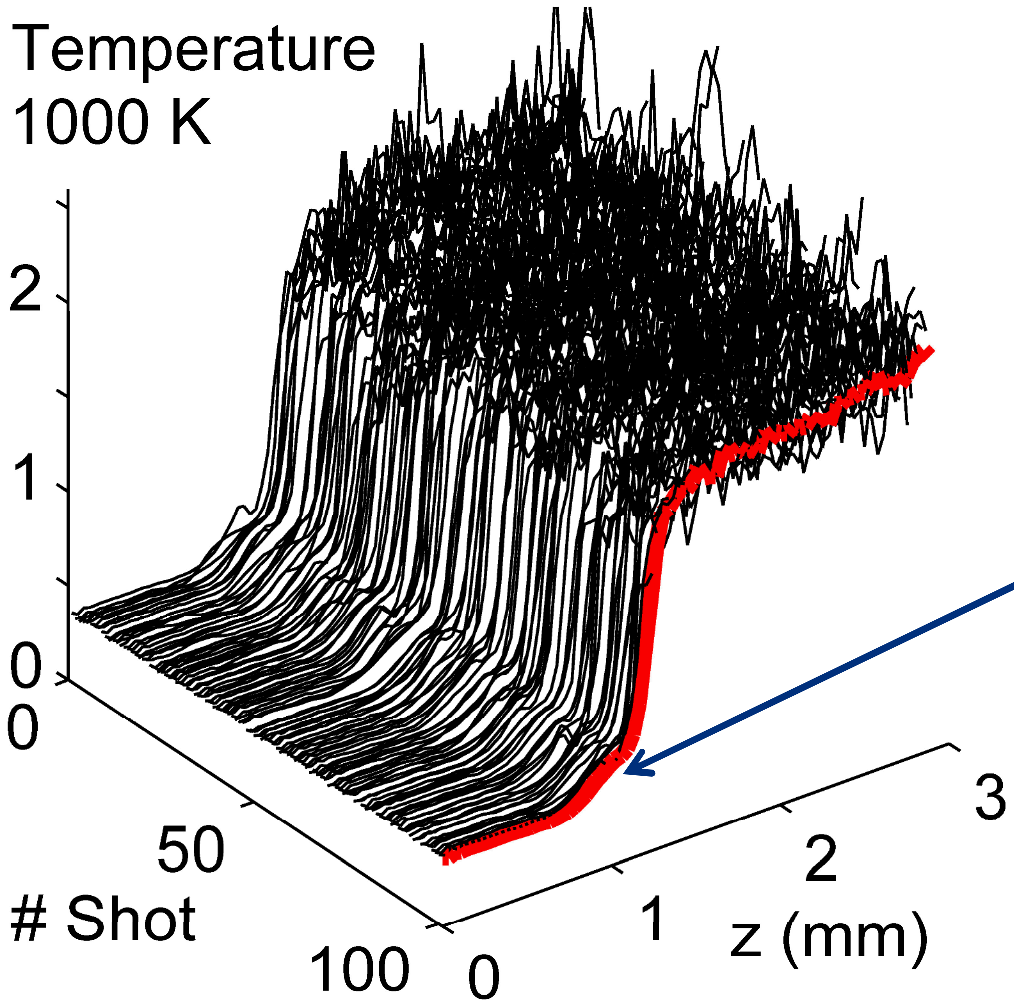
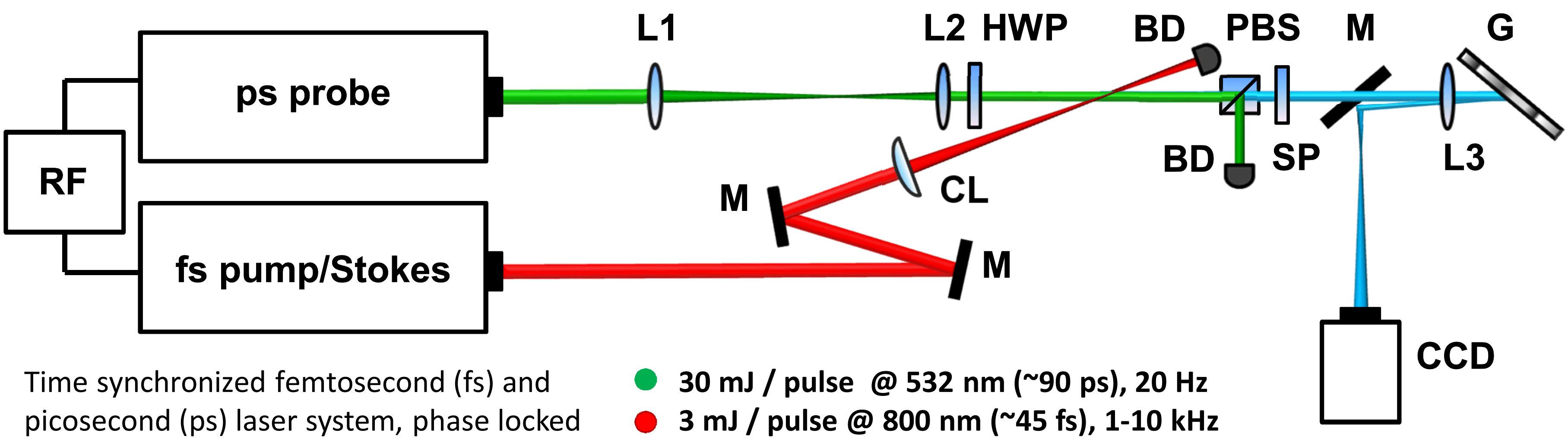


Excellent spatial resolution may reveal interesting new features…

…or is it just beam steering…

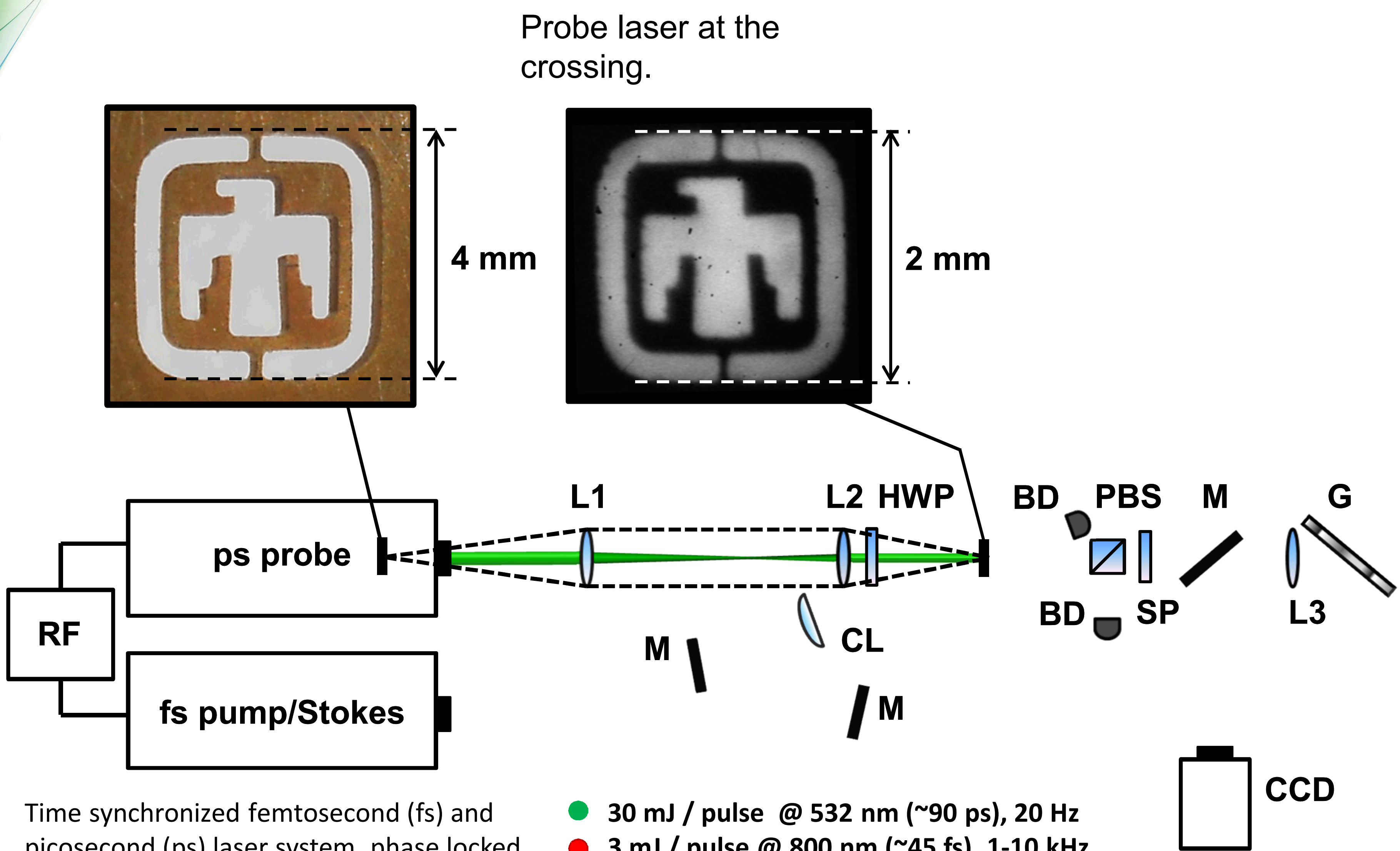
Spectrally resolved detection of 2D-CARS



Time synchronized femtosecond (fs) and picosecond (ps) laser system, phase locked to an external 100 MHz RF source.

- **30 mJ / pulse @ 532 nm (~90 ps), 20 Hz**
- **3 mJ / pulse @ 800 nm (~45 fs), 1-10 kHz**

Spectrally resolved detection of 2D-CARS



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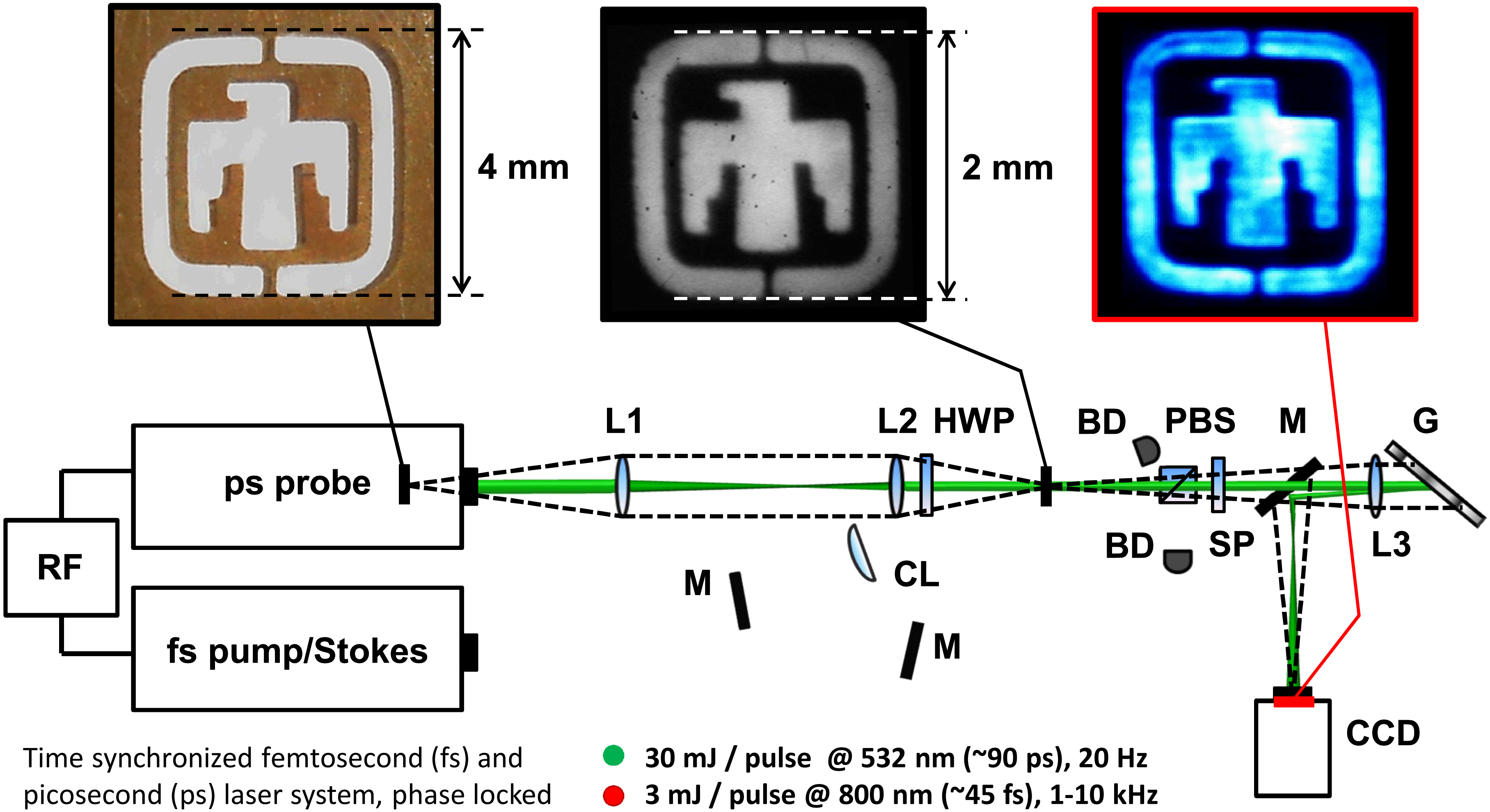
Spectrally resolved detection of 2D-CARS

Probe laser at the crossing.

Small portion of the probe light imaged through a grating.



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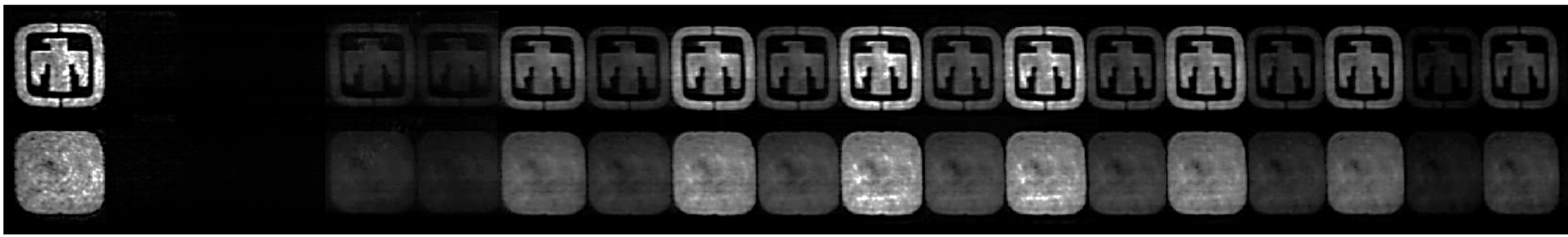
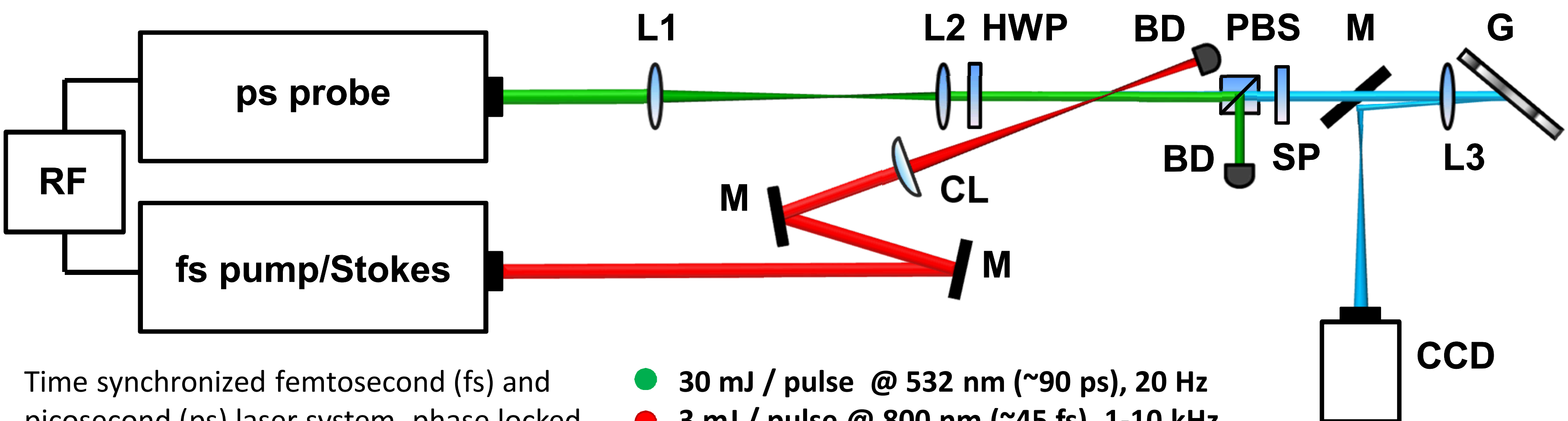
Spectrally resolved detection of 2D-CARS

Rotational quantum number *J* = 4 5 6 7 8 9 10 11 12 13 14 15 16



120 X 125 pixels = 15000 spatially correlated spectra in a single laser shot.



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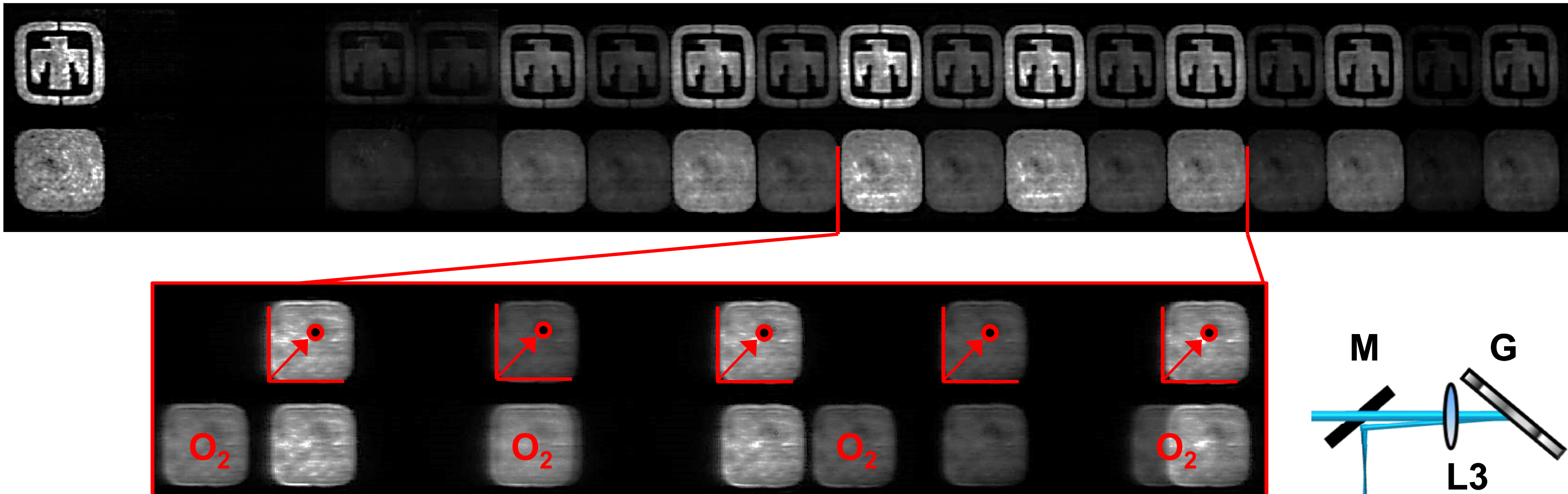
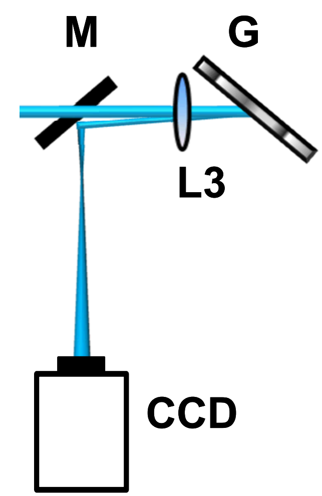
Simultaneous planar imaging and multiplex spectroscopy in a single-shot

Rotational quantum number *J* = 4 5 6 7 8 9 10 11 12 13 14 15 16



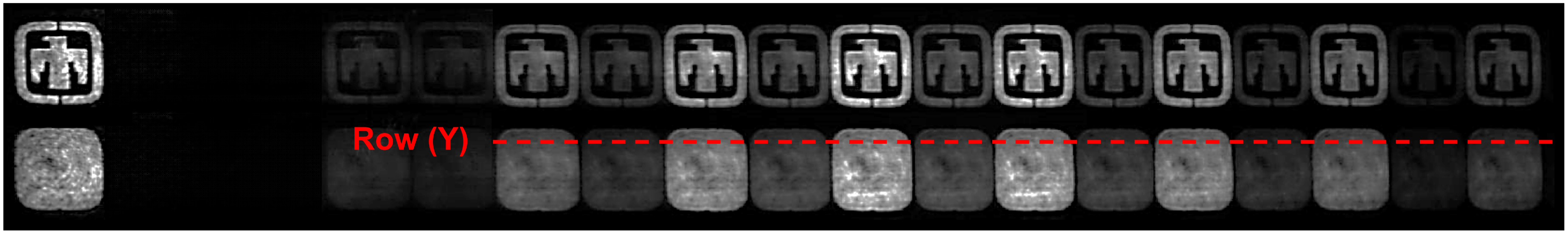


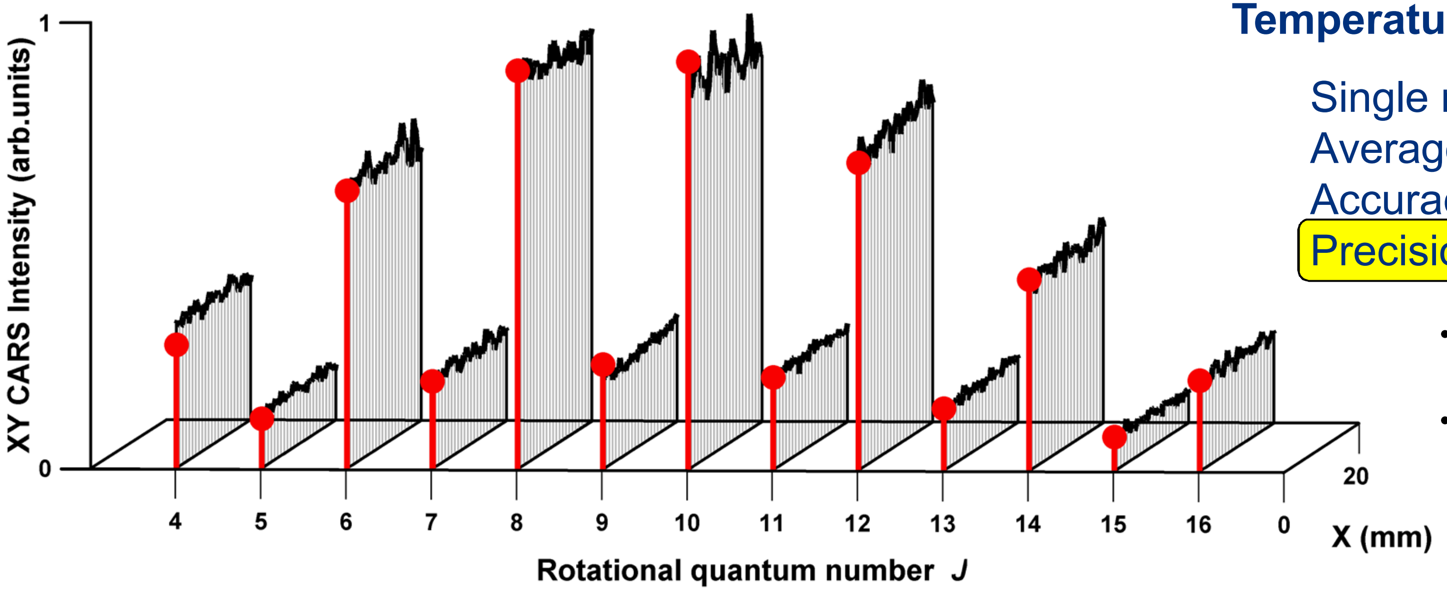
- Tunable spectral dispersion, enabling multispecies detection and probing of a larger 2D field.
- Vector diagram to orientate each location of the spatially resolved measurement.

B. K. Ford, *et al.*, *Opt. Express* **9**, 444 (2001).

Simultaneous planar imaging and multiplex spectroscopy in a single-shot

Rotational quantum number *J* = 4 5 6 7 8 9 10 11 12 13 14 15 16





Temperature field statistics

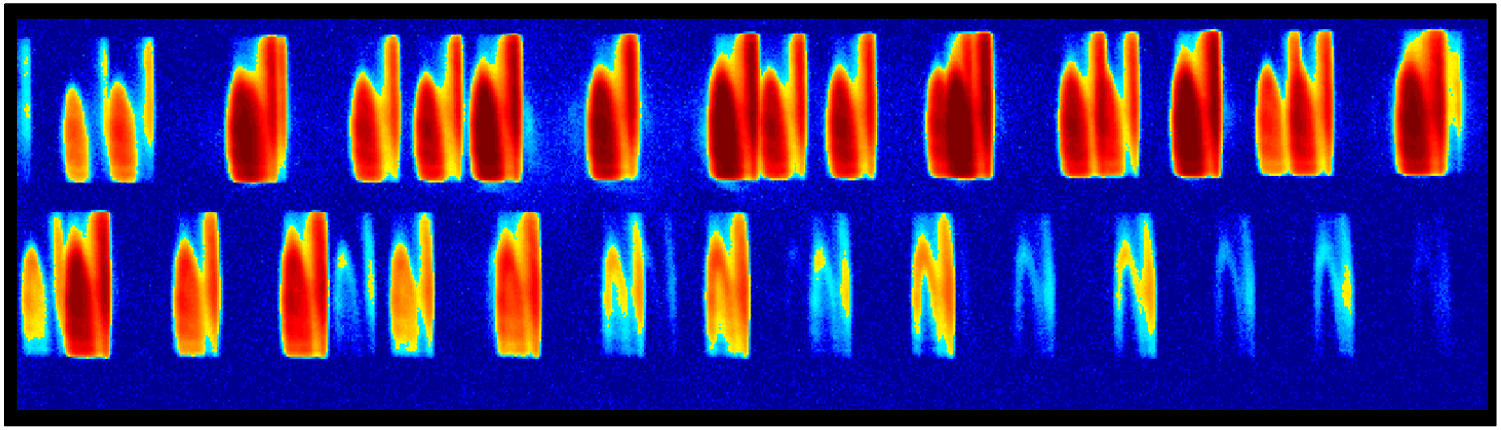
Single row ~ 120 spectra
Average T = 299.6 K
Accuracy = 1.5 %
Precision = 1.4 %

- **Pixel-to-pixel extraction of spectra.**
- **Insensitive to irregularities in the probe & excitation pulses spatial profiles.**

2D-mapping of temperature and species in flames

- Detecting #25 N_2 and #14 O_2 S-branch transitions with small spectral interference.

N$_2$ S(14) @ 123.18 cm^{-1}



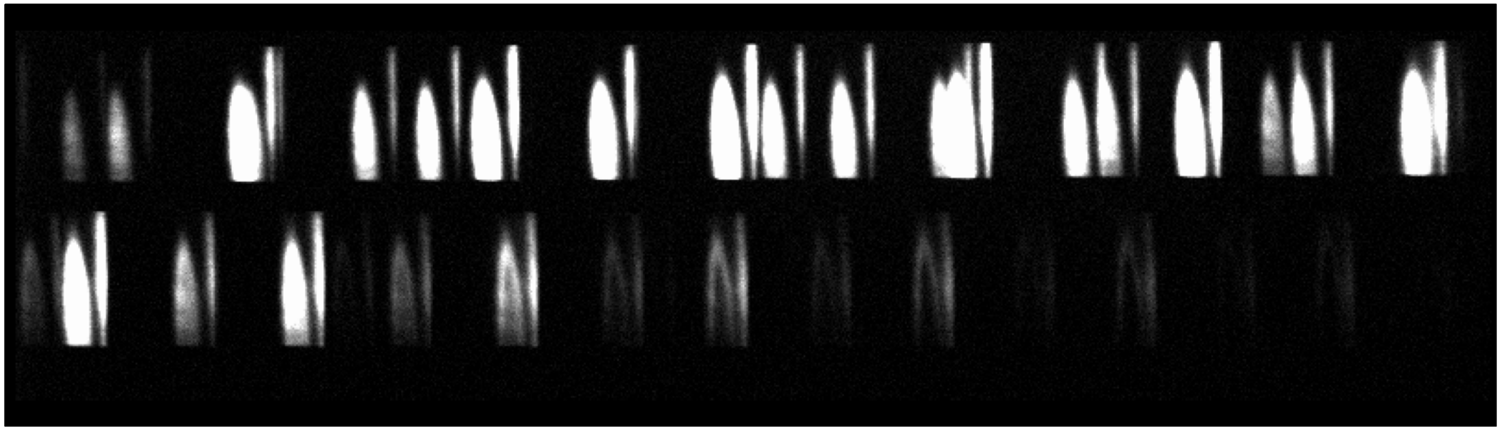
100 accumulated shots

- The measurements are optimized for flame thermometry and detecting $[N_2]/[O_2]$, i.e. narrower mask, ~2100 spectra collected simultaneously, 2D-field of 2 x 7.5 mm.

2D-mapping of temperature and species in flames

- Detecting #25 N_2 and #14 O_2 S-branch transitions with small spectral interference.

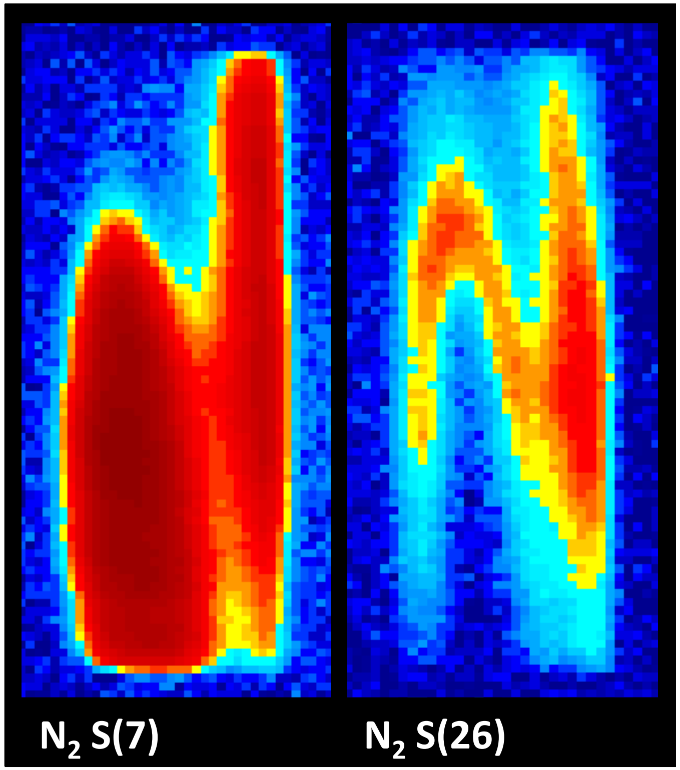
N_2 S(14) @ 123.18 cm^{-1}



Single shot raw data collected @ ~5 Hz

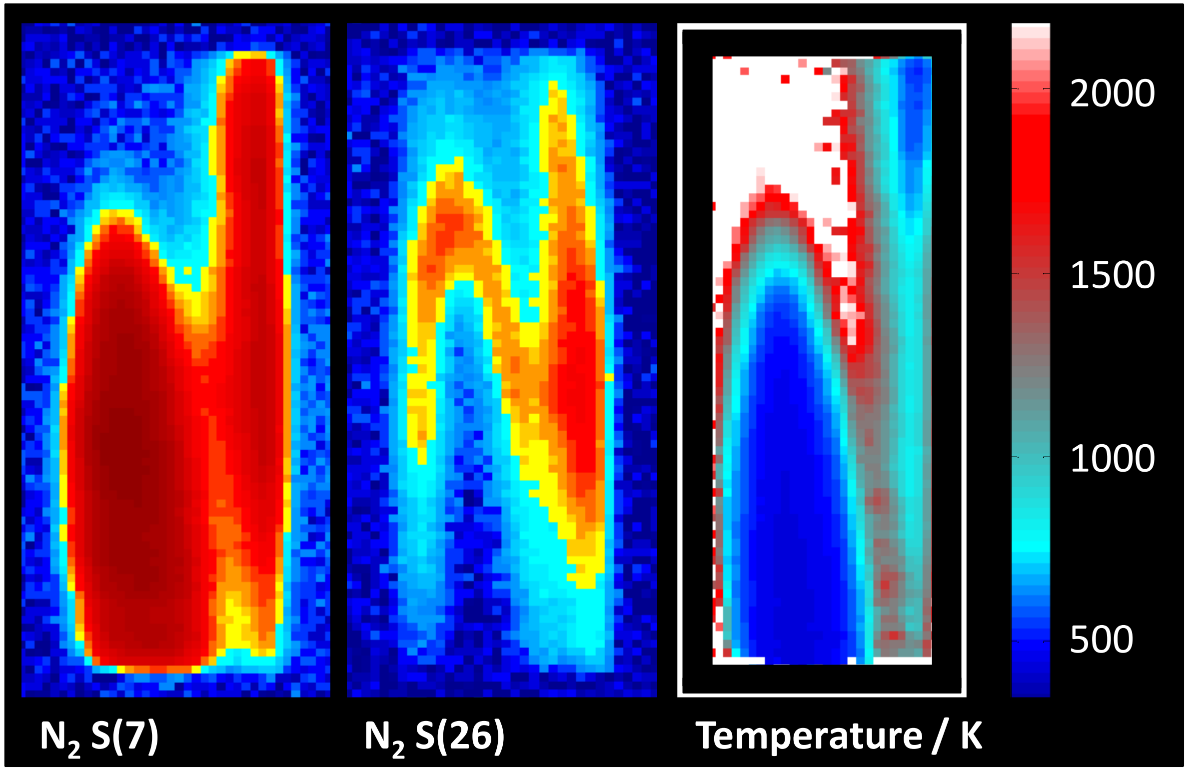
- The measurements are optimized for flame thermometry and detecting $[N_2]/[O_2]$, i.e. narrower mask, ~2100 spectra collected simultaneously, 2D-field of 2 x 7.5 mm.

2D-mapping of temperature and species in flames



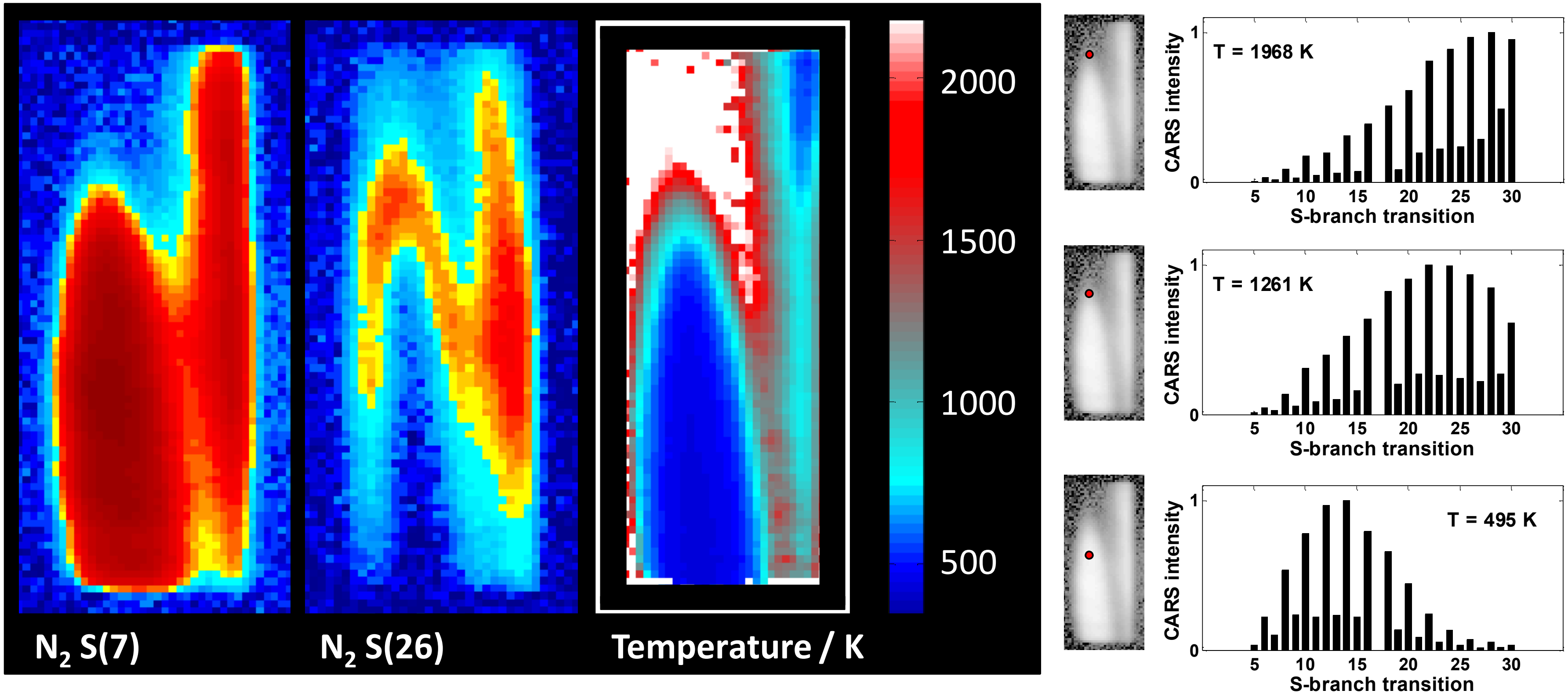
- Imaging the spatial distribution of cold and hot N_2.

2D-mapping of temperature and species in flames



- Imaging the spatial distribution of cold and hot N_2.
- Temperatures extracted in the range from 300K – 2000K.

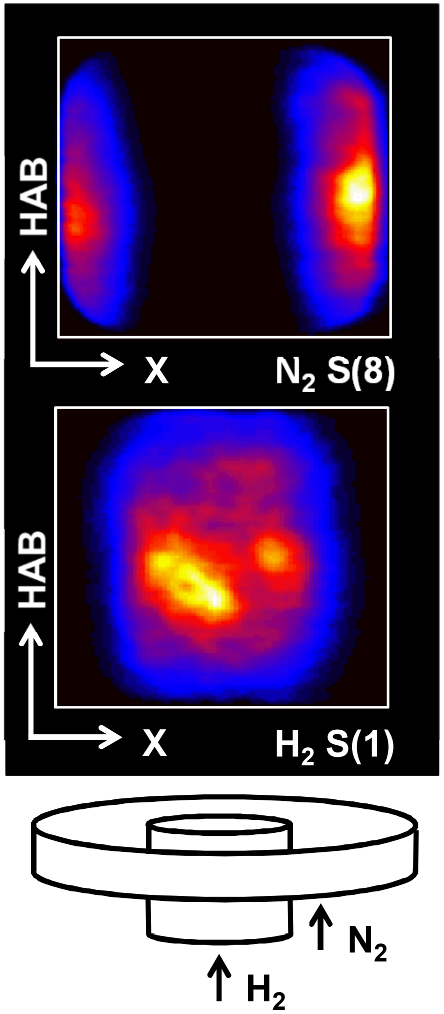
2D-mapping of temperature and species in flames

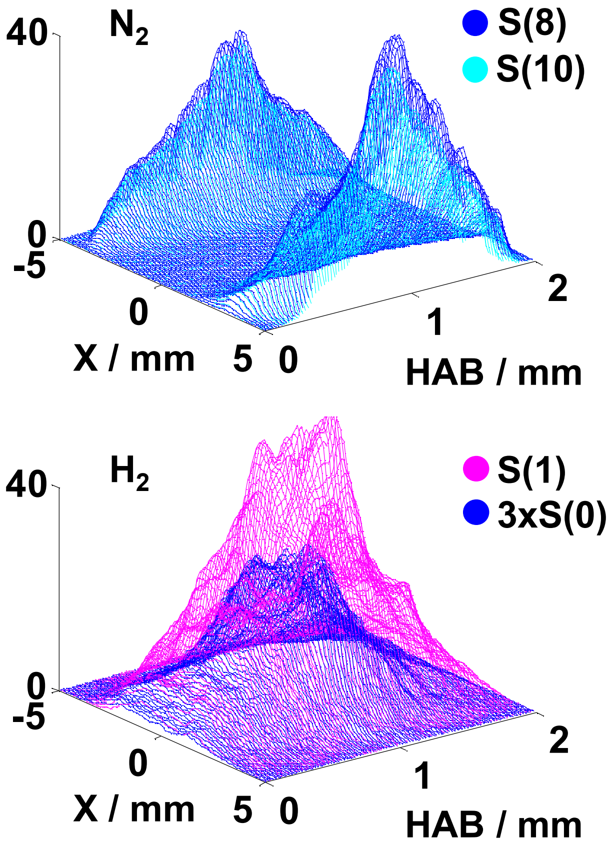


- Imaging the spatial distribution of cold and hot N_2.
- Temperatures extracted in the range from 300K – 2000K.

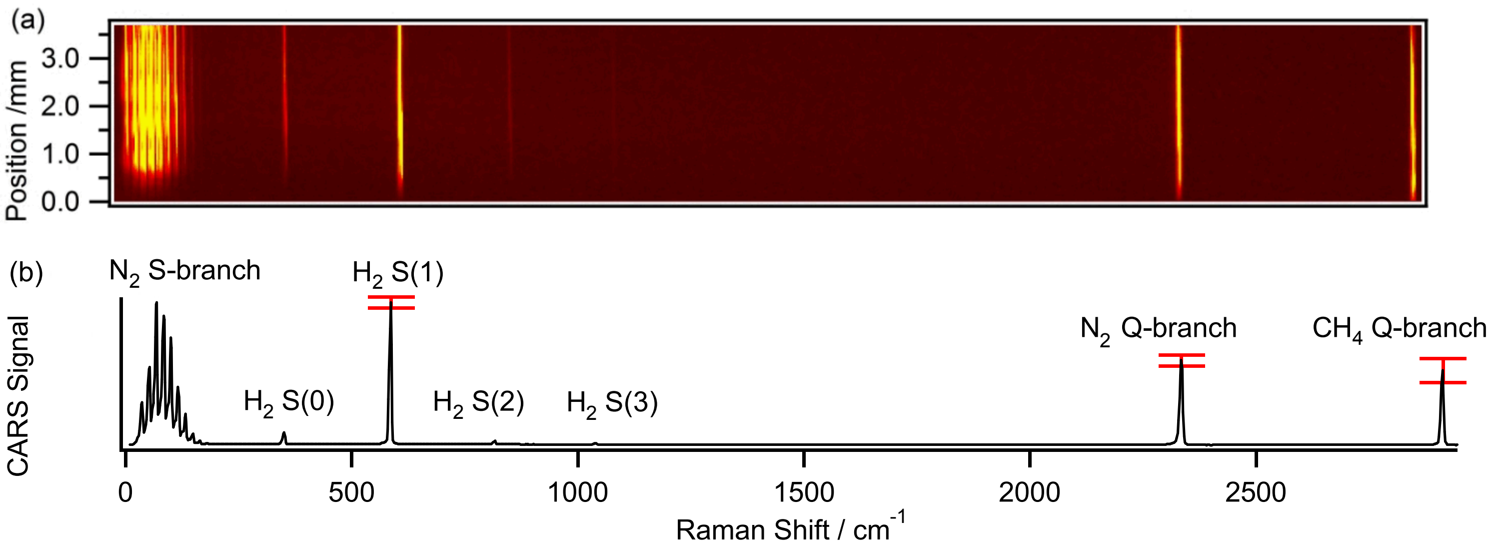
2D-mapping of temperature and species in flames

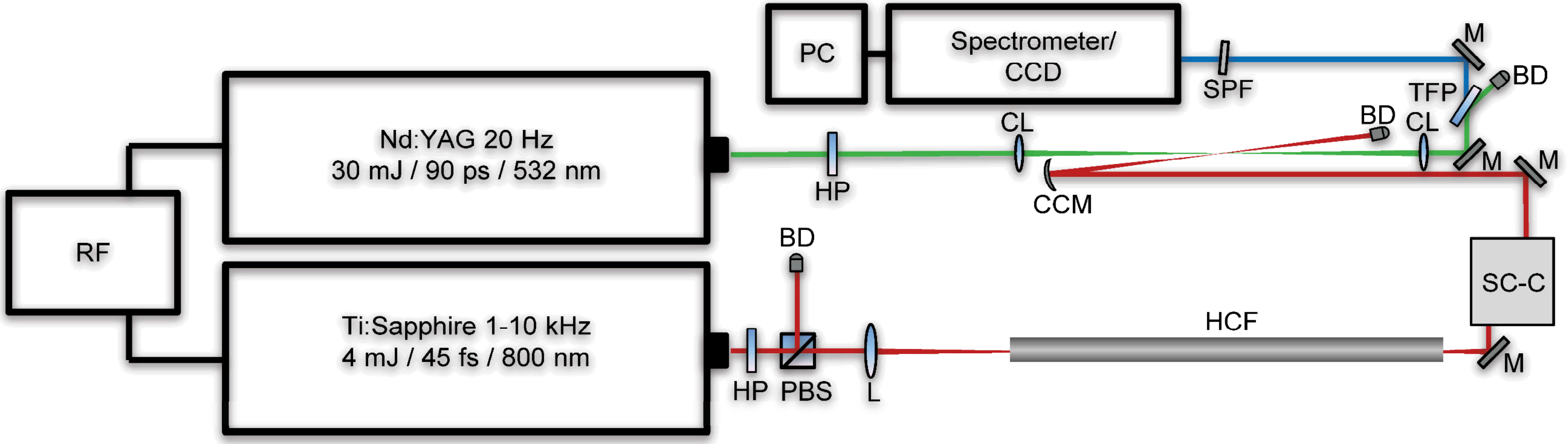
- Two-dimensional single-shot imaging of a H_2 jet-stream co-flowed with N_2
- Inhomogeneity in the excitation and probe laser profile is repeated in all the spectral transitions, 2D-CARS is self-referenced against these effects.



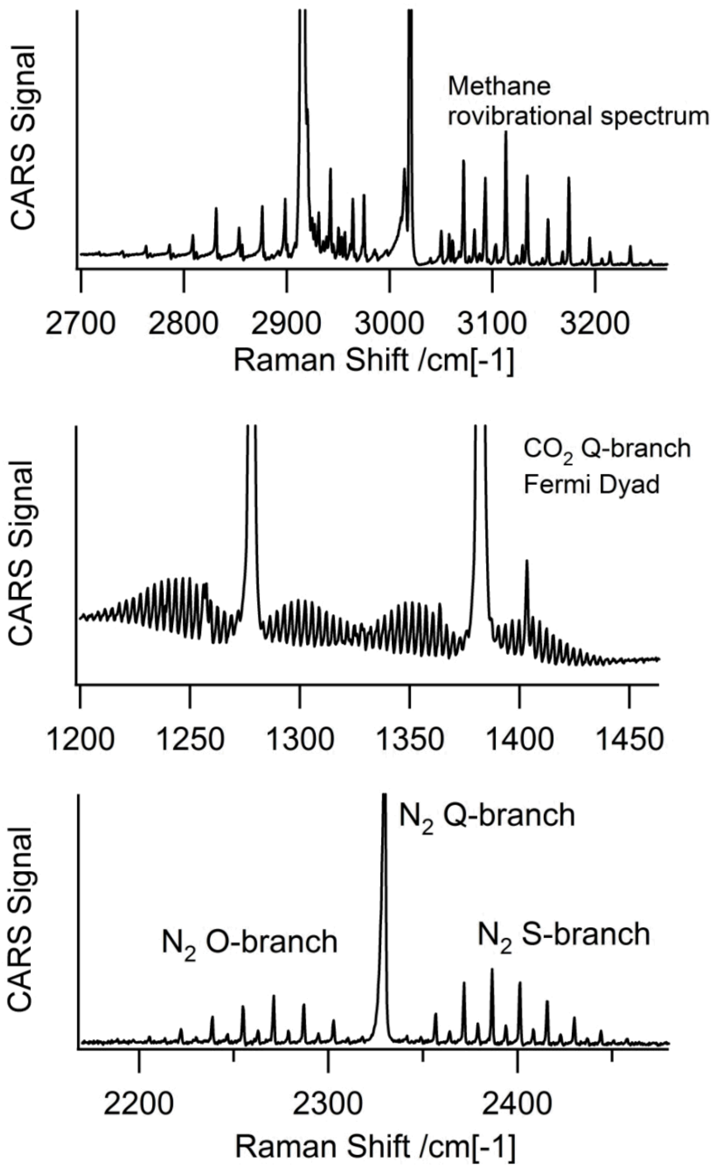


Two-beam Ultrabroadband CARS

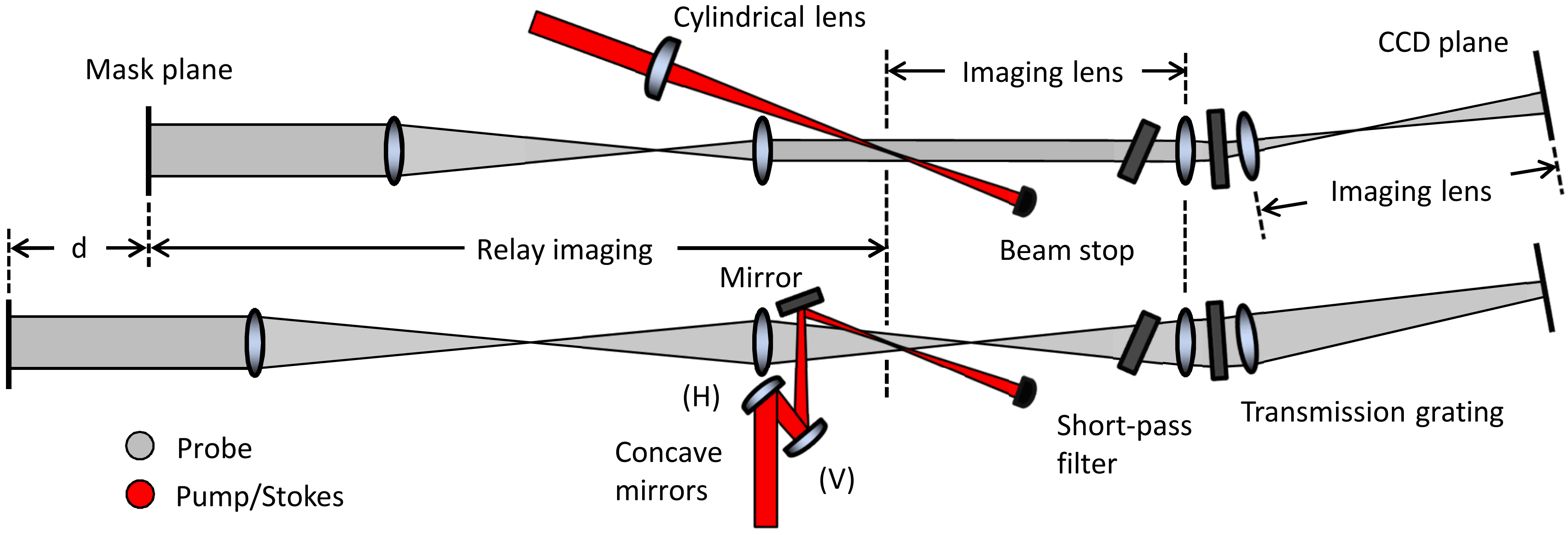


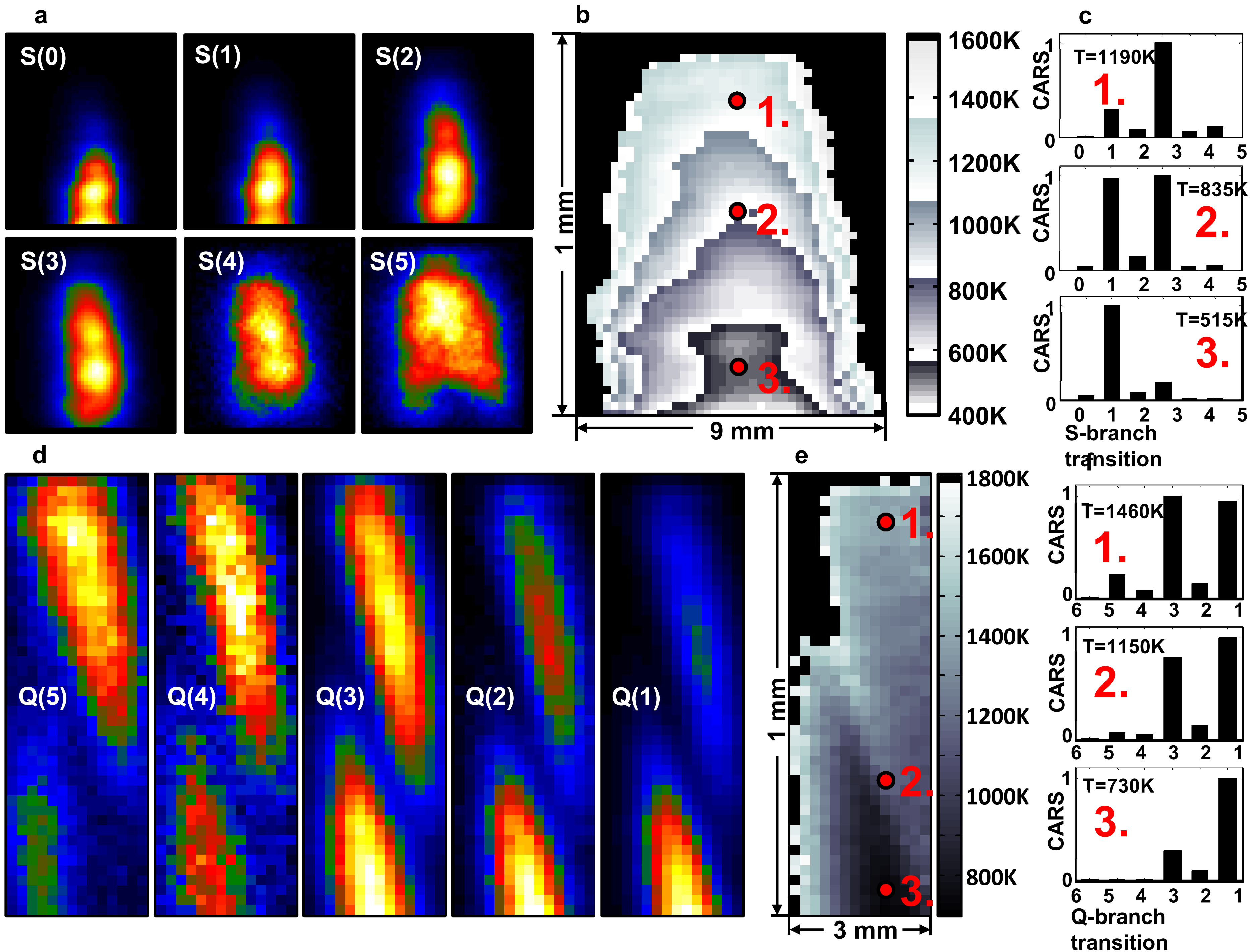


Two-beam Ultrabroadband CARS



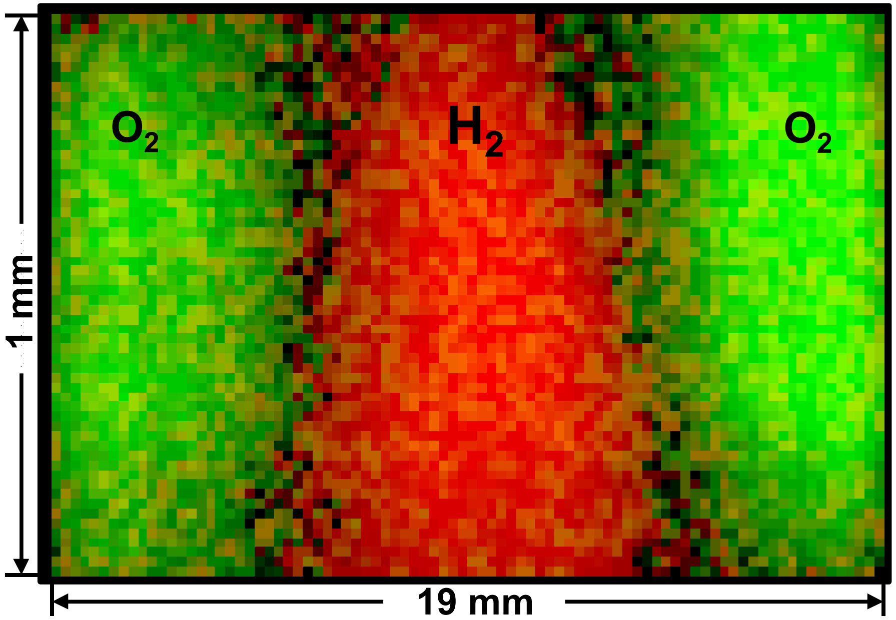
Two-beam Ultrabroadband CARS





a
S(0)
S(1)
S(2)
S(3)
S(4)
S(5)
b
1 mm
9 mm
1.
2.
3.
1600K
1400K
1200K
1000K
800K
600K
400K
c
T=1190K
T=835K
T=515K
CARS
S-branch transition
d
Q(5)
Q(4)
Q(3)
Q(2)
Q(1)
e
1 mm
3 mm
1800K
1600K
1400K
1200K
1000K
800K
f
T=1460K
T=1150K
T=730K
Q-branch transition

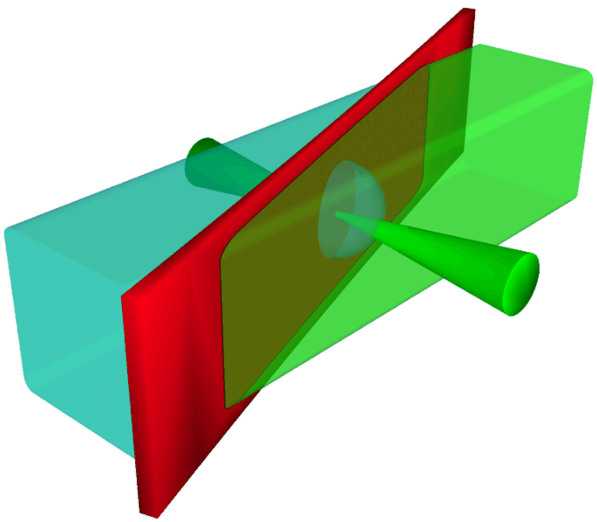
Two-beam Ultrabroadband CARS



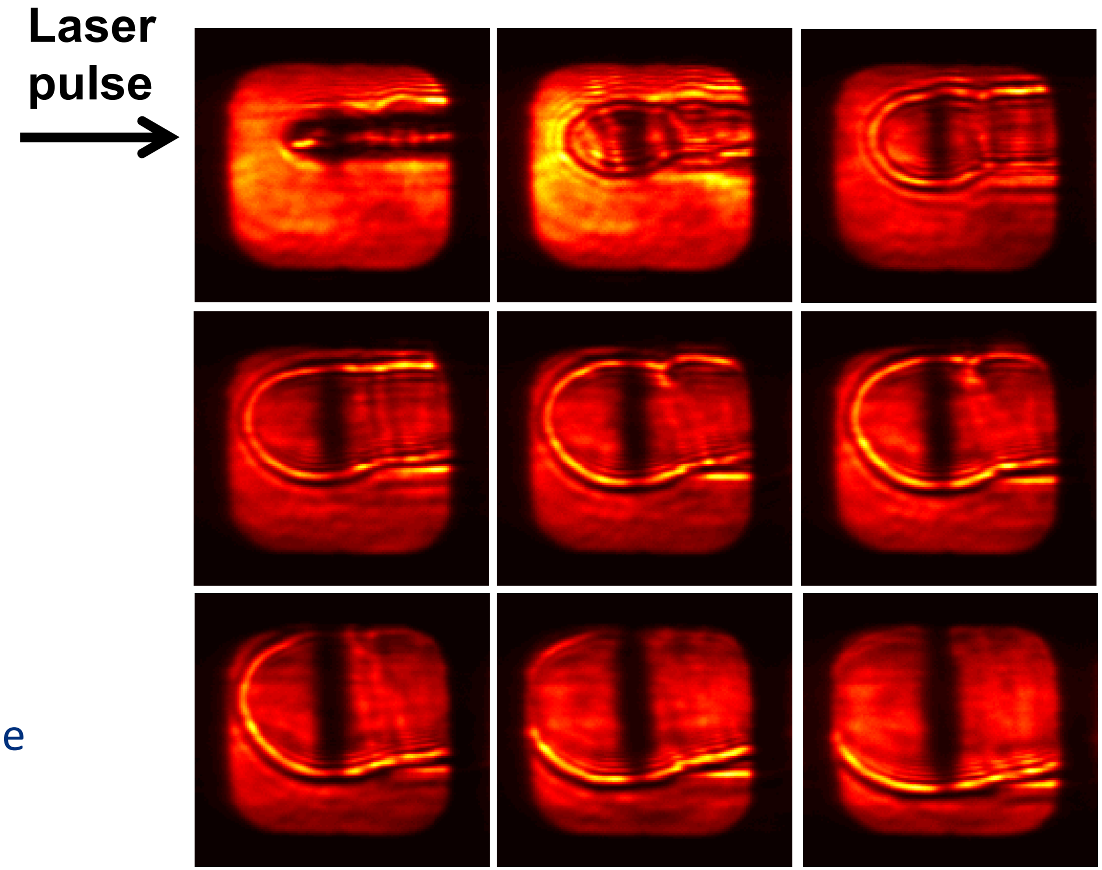
- Direct CARS imaging of fuel / oxidizer
- Thermometry directly on fuel molecule

Preliminary low temperature application in intense laser pulse induced plasma

Measurement geometry



Plasma – CARS delay, 15ns – 175ns



- Time-resolved measurements studying the propagation of the plasma induced shock wave.
- Observed rotational heating.