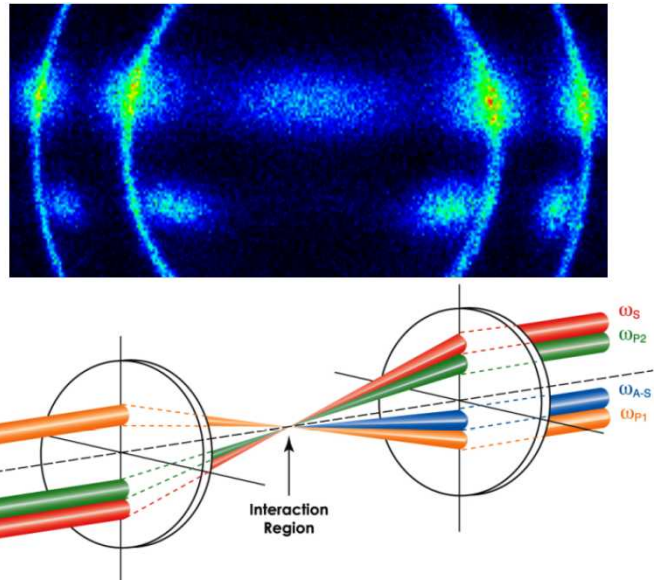
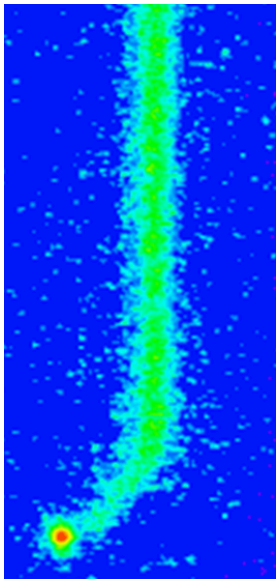
Molecular-based optical measurement techniques for aerospace flows

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Supported by the NASA Fundamental Aeronautics Program: High Speed Project

Outline

- Measurements for Aerospace Flows
  - Introduction
  - What makes a good measurement system?
  - Measurement Requirements
  - Technique Overviews

  } Lecture 1

- Planar Laser-Induced Fluorescence (PLIF) } Lecture 2

- Rayleigh Scattering, Raman Scattering and Coherent anti-Stokes Raman Spectroscopy (CARS) } Lecture 3

Aerospace Flows

- Aerospace flows and flow facilities are wide ranging:



http://www.wpafb.af.mil/news/story.asp?id=123181921



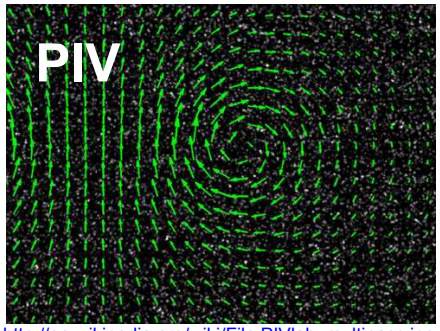
http://www.nasa.gov/centers/langley/multimedia/iotw-ntf-model.html

Micro air vehicles ←→ Commercial transport aircraft

- ***Reynolds numbers***: low (laminar) to high (turbulent)
- ***Mach numbers***: low (incompressible) to high
  - Near Mach 1 and higher: compressibility, variable temperature, shock waves
  - High Mach: large P, T changes, chemical reactions
- Any of these flow regimes can have combustion

Measurement Techniques

- Typical parameters of interest:
  - Velocity, Pressure, Density, Temperature, Species
- No one measurement technique is suitable for studying all these different flow phenomena
- Consider low-Mach number flows:
  - Velocity is of primary interest
  - Density ~ constant
  - Can infer pressure from velocity field
  - Often can use Particle Image Velocity (PIV) or Laser Doppler Velocimetry (LDV) to get most required information
  - If that works → use it!
    - PIV, LDV are generally accurate, precise, commercially available, straight forward to implement, widely used, widely accepted.
    - Successfully applied at Mach 5 (UT Austin), even Mach 8 (Princeton)! But in these flows, PIV doesn't measure everything.



http://en.wikipedia.org/wiki/File:PIVlab_multipass.jpg

Some Limitations of PIV / LDV

- In many flows PIV and LDV *cannot* provide the required information:
  - If density, temperature, pressure, species concentrations are needed
  - If particles cannot be seeded into the flow
  - If particles do not follow the flow (vortex, shocks)
  - If particles coat the windows (high temperature)
  - If particles do not survive or interact with flow
- In these cases, *spectroscopic* measurement techniques can provide the required information in Aerospace Flows
  - Topic of this week's lecture series

Some advantages of Spectroscopic Measurement Techniques

- Non-intrusive
  - No probes, particles perturb the flow
- Remote detection
  - Can be used in hostile environments
- Can measure multiple parameters,
  - Sometimes simultaneously
  - Can be instantaneous (<10 ns) and high freq. (MHz)
  - Can be precise, accurate, high spatial res. (2D, 3D)
- *But…*
  - Can have more complicated theory, expensive and complex equipment, more difficult data analysis
  - May require seeding of (sometimes toxic) gases, more expertise, slower data acquisition, analysis

Scope of the Content

- In both the introductory material and the content presented later in the week, we focus on molecular based, quantitative, mostly spectroscopic measurement techniques applied in or applicable to aerospace facilities.

Will emphasize our team's work

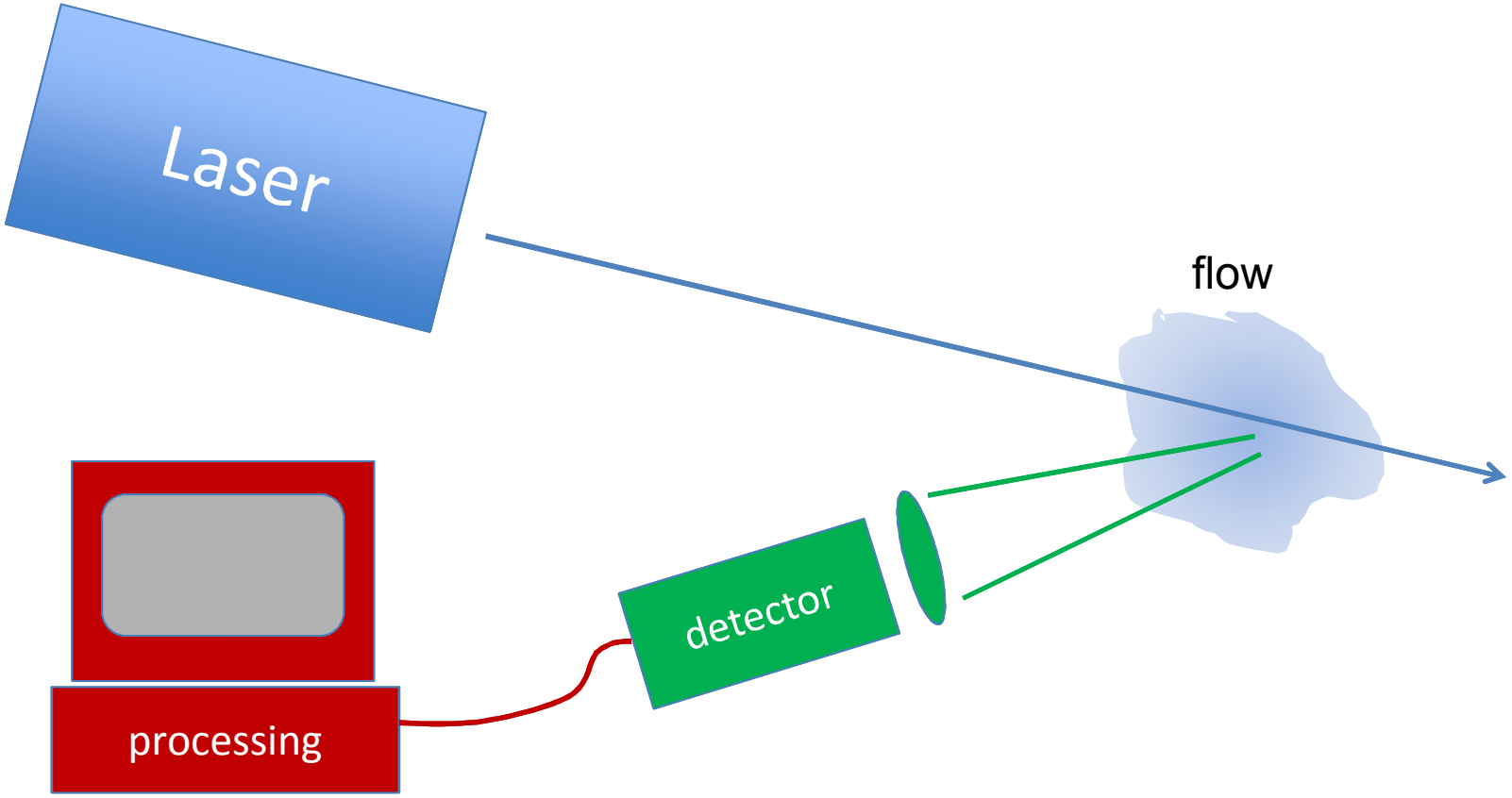
- *All lectures <u>exclude</u> the many highly relevant particle-based, probe-based and surface based measurement techniques for high speed flow; mostly no 'flow vis'.*

What makes a good measurement technique?

- A reliable measurement system matched with well-understood physics to make a quantified measurement that meets a customer's requirements.

  Explore each of these…

Typical Measurement System



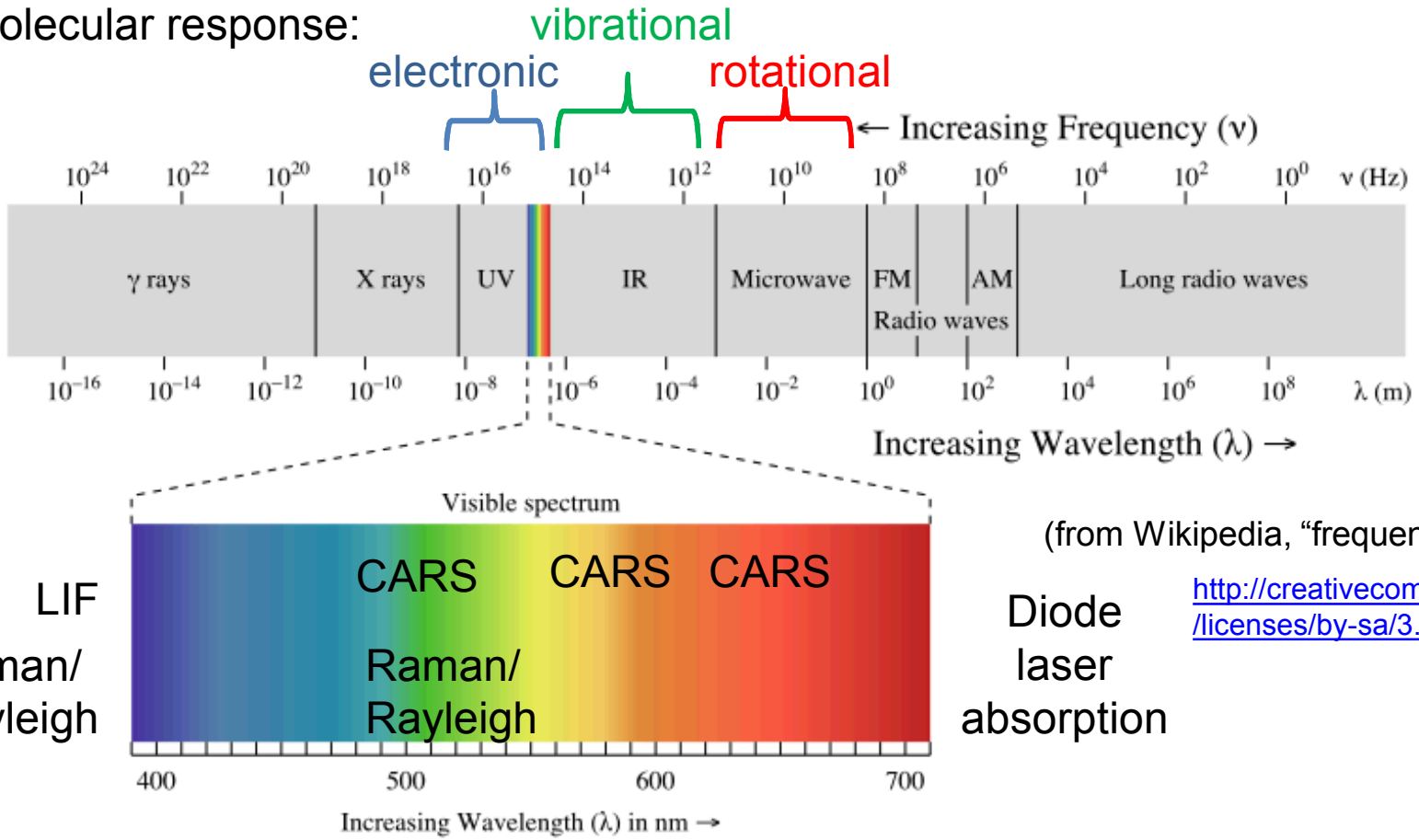
- Need to understand equipment and physics of process to make a measurement
  - Techniques: LIF, Rayleigh, Raman, CARS, etc.

Measurement System 1

- Lasers: pulsed vs. continuous wave (cw)
  - Continuous wave: lower power (mW to Watt)
  - Pulsed:
    - 10 ns, picosecond, femtosecond all commercially available
    - ‘freeze’ the flow
    - much higher power (MWatt-GWatt, even TWatt)
    - Frequency conversion (useful in linear techniques such as Raman, Rayleigh, LIF)
    - Provide high powers for nonlinear techniques (CARS)
    - Repetition rate: 10-100 Hz, 100 kHz, 1 MHz, 80 MHz…
- What colors of laser light do we need?

Colors of Light: Spectroscopy

Molecular response:



LIF

Raman/ Rayleigh

CARS CARS CARS

Raman/ Rayleigh

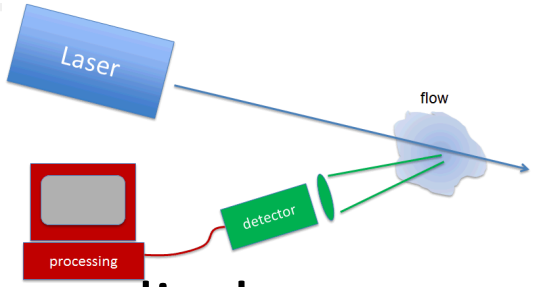
Diode laser absorption

(from Wikipedia, "frequency")

http://creativecommons.org/licenses/by-sa/3.0/deed.en

- Many different colors can be useful for spectroscopy
- As energy increases, more modes can be excited
  - LIF: can see electronic, vibrational and rotational structure

Measurement System 2

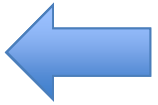


- Detectors:
  - Single point detectors:
    Photomultipler tubes (PMT) and photodiodes
  - Camera technology is rapidly evolving:
    - CCDs provide ‘scientific grade’ (low noise, linear, etc.)
    - CMOS cameras are faster
    - Commercially available cameras at up to 10’s, 100’s of kHz steady streaming or MHz for short bursts or few pix
  - Fiber optics and fiber-optic bundles often used for either transmitting laser or collecting signal
- Processing:
  - Some data processing codes are available (CARSFT, LIFBASE) but many are proprietary.

What makes a good measurement technique?

- A reliable measurement system matched with well-understood physics to make a quantified measurement that meets a customer's requirements.



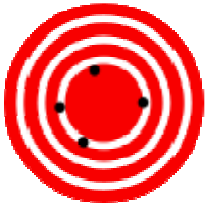


Explore each of these…

Quantified Measurements

- Accuracy and precision of a measurement must be quantified to be useful, for example when comparing to a code
  - Qualitative measurements (flow vis) can be useful

- Quantified measurement:
  - Accuracy and precision are well characterized



Accurate but not precise



Precise but not accurate

(from Wikipedia, "accuracy and precision")

  - Accuracy: compared to a 'standard' or 'accepted'
  - Precision: 1σ of large sample of measurements

Customer Requirements

- Consider a customer with arbitrary measurement requirements:
  - Would like a particular parameter measured
  - Would like the measurement at some conditions (P, T)
    - Many measurement technique signals scale as P_{static}, T_{static}
- Parameters to be measured:
  - Velocity, temperature, density, pressure, species concentration or mole fraction
- Should base choice of measurement technique on prior success by others
  - Have compiled past work into graphics that show static pressure and temperature of demonstrated measurements
  - Received contributions from other authors
  - Use these charts to find appropriate measurement methods

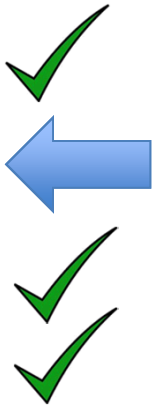
Additional Customer Requirements

Interview the customer… (sometime the customer is you!)

- Simultaneous measurements to obtain correlations: T'u', ρu'v'
- Time resolution (< μsec), and frequency response (> MHz)
- Spatial resolution: near walls; smallest scales in shear layer
- Where are the measurement to be made?
  - Inflow to establish B.C., exit, near walls?
  - Point, line, plane, volume?
- Accuracy and Precision Requirement
- What quantity of data is required?
- When is the data needed? Is "real-time" data required?
- What type of optical access is available?
- Can (toxic) seed gases be introduced? Will they influence the properties being measured?
- What is the ordered priority of the above requirements?

What makes a good measurement technique?

- A reliable measurement system matched with well-understood physics to make a quantified measurement that meets a customer's requirements.



- Now the lecture series will focus on different measurement systems, introducing the physics.
  - How's the basic physics of the method work?
  - Can the measurements be made instantaneously (aka. single-shot) at high frequencies?
  - What accuracy and precision can be obtained?

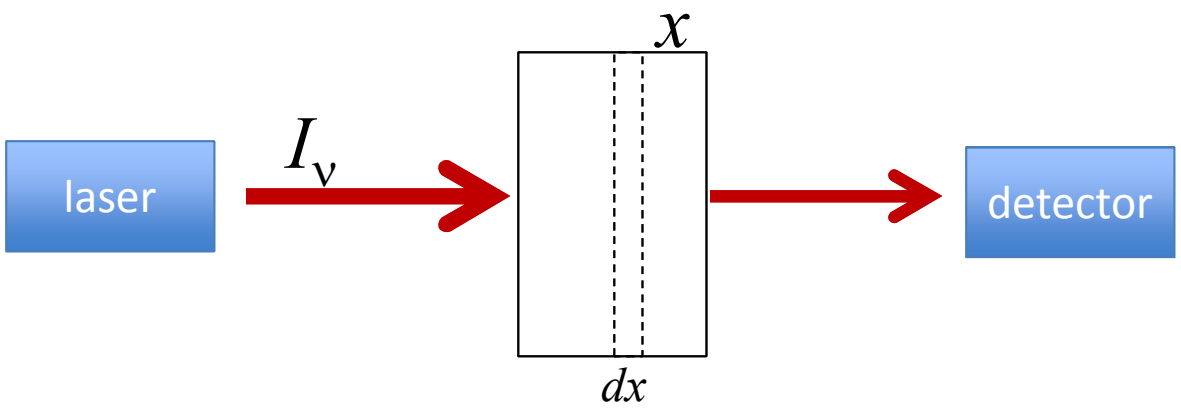
Various optical or spectroscopic measurement techniques for aerospace flows (short descriptions)

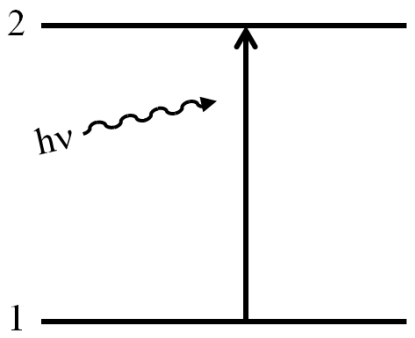
Selected “Other” techniques

- (PLIF, Rayleigh, Raman, CARS described in detail later this week.)
- Describe a few recent techniques of interest for measuring in aerospace flows
  - Tunable Diode Laser Absorption Spectroscopy (TDLAS)
  - Background Oriented Schlieren (BOS)
  - Degenerate Four-Wave Mixing (DFWM)
  - Laser-Induced Thermal Acoustics (LITA)

- Something in common:
  - These technique can be used to study turbulent flows, but generally cannot resolve small turbulent structures

Tunable Diode Laser Absorption Spectroscopy





- Absorption is described by Beer-Lambert Law:

$$-dI_\nu = I_\nu k_s(\nu) dx$$

where $k_s(\nu)$ is the spectral absorption coefficient.

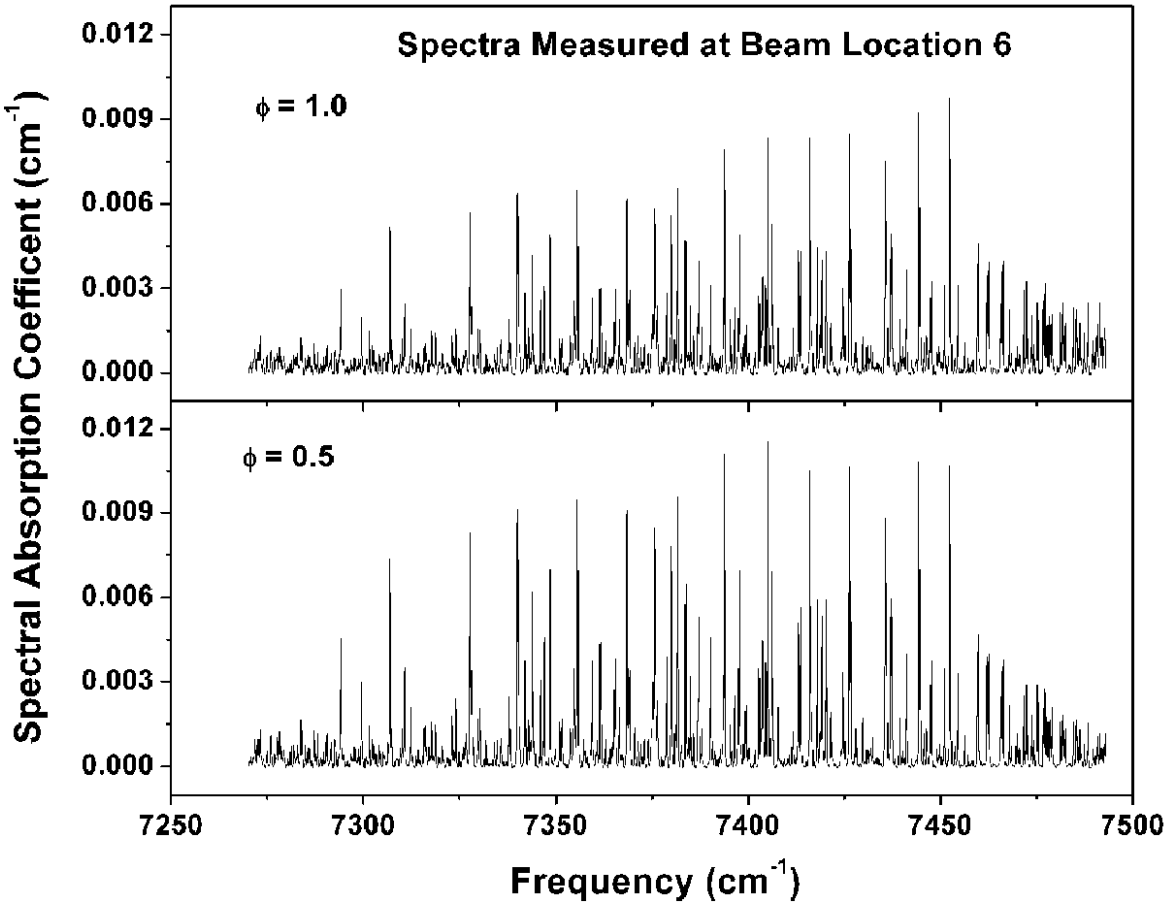
- This leads to exponential decay of laser intensity:

$$I_{\nu,x} = I_{\nu,0} e^{-k_s(\nu)x}$$

- Measure I_ν, know x, directly measure $k_s(\nu)$
- $k_s(\nu)$ contains information about temperature, concentration, velocity of gas
- Problem: path averaged line-of-sight technique

Tunable Diode Laser Absorption Spectroscopy

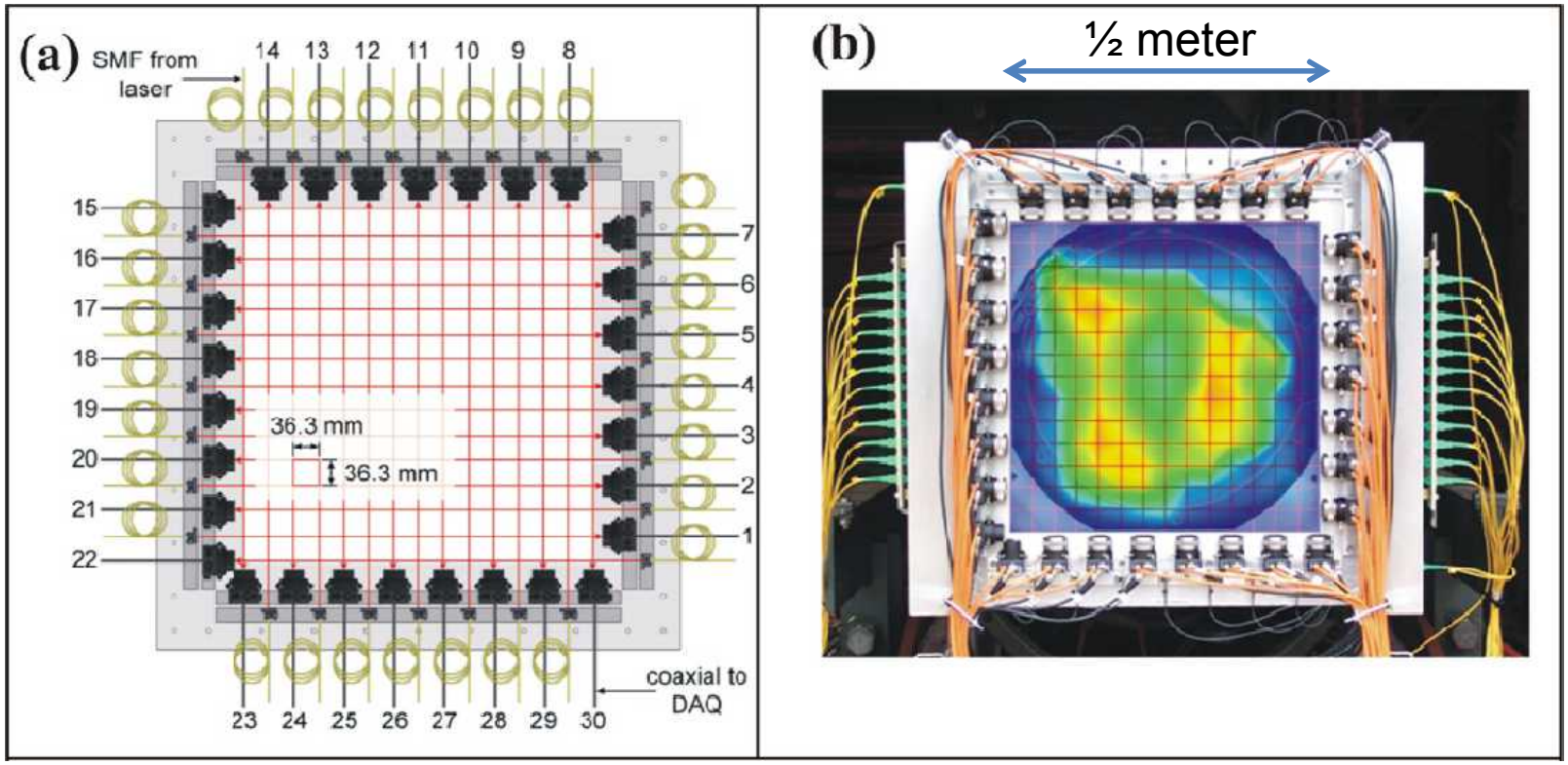
- Can measure $k_s(\nu)$ over a single line or many lines:



L. Ma, W. Cai, A. W. Caswell, T. Kraetschmer, S. T. Sanders, S. Roy, and J. R. Gord, "Tomographic imaging of temperature and chemical species based on hyperspectral absorption spectroscopy," Optics Express 17(10), 8602–8613, 2009.

TDLAS Tomography

- Major limitation of TDLAS: path-averaged
  - Can do tomography

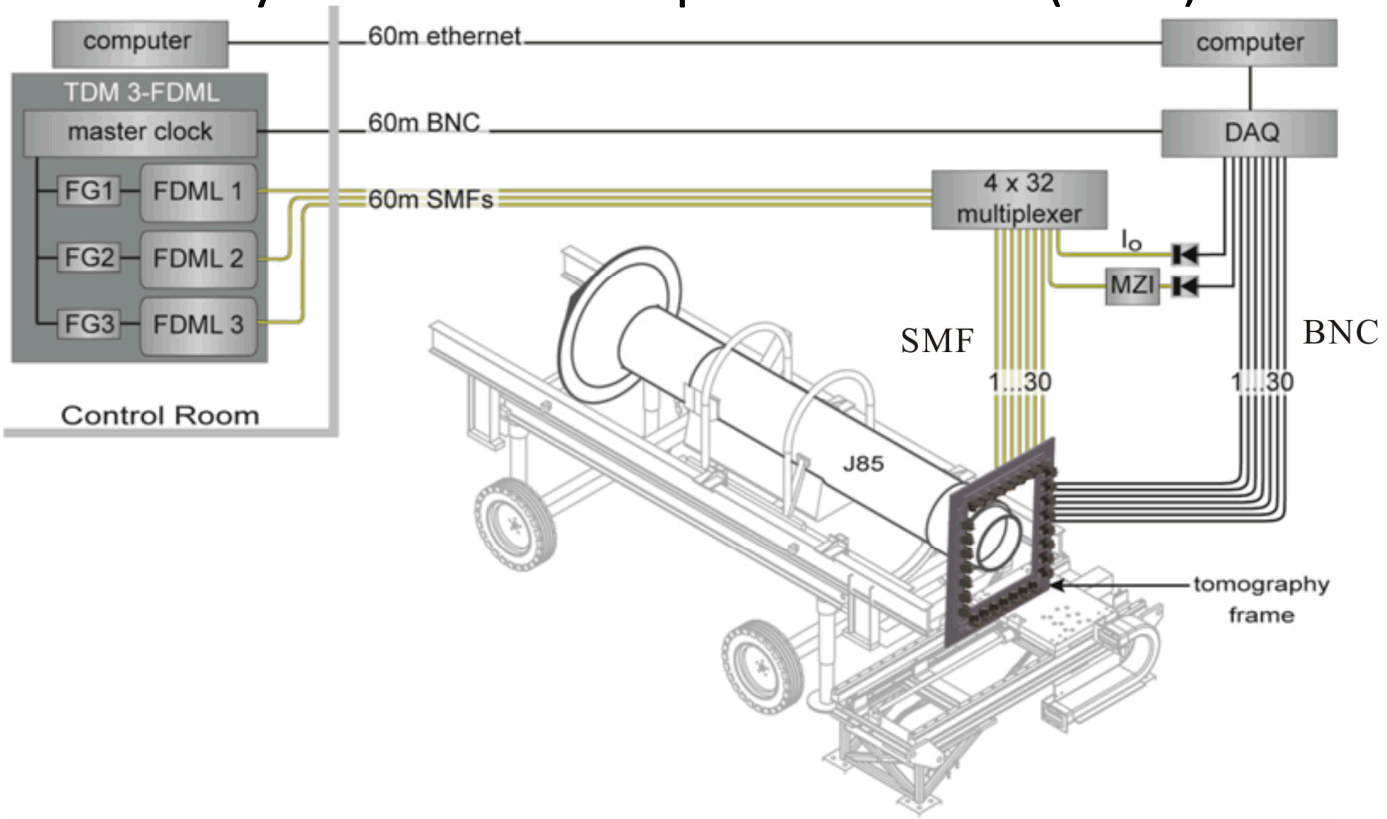


  - 15x15 grid provides data at 225 points (36 mm grid)

L. Ma, X. Li, S. T. Sanders, A. W. Caswell, S. Roy, D. H. Plemmons, and J. R. Gord, "50-kHz-rate 2D imaging of temperature and H_2O concentration at the exhaust plane of a J85 engine using hyperspectral tomography," Optics Express, Vol. 21, Issue 1, pp. 1152-1162, 2013

TDLAS Tomography

- General Electric J85 gas turbine engine
  - University of Tennessee Space Institute (UTSI)

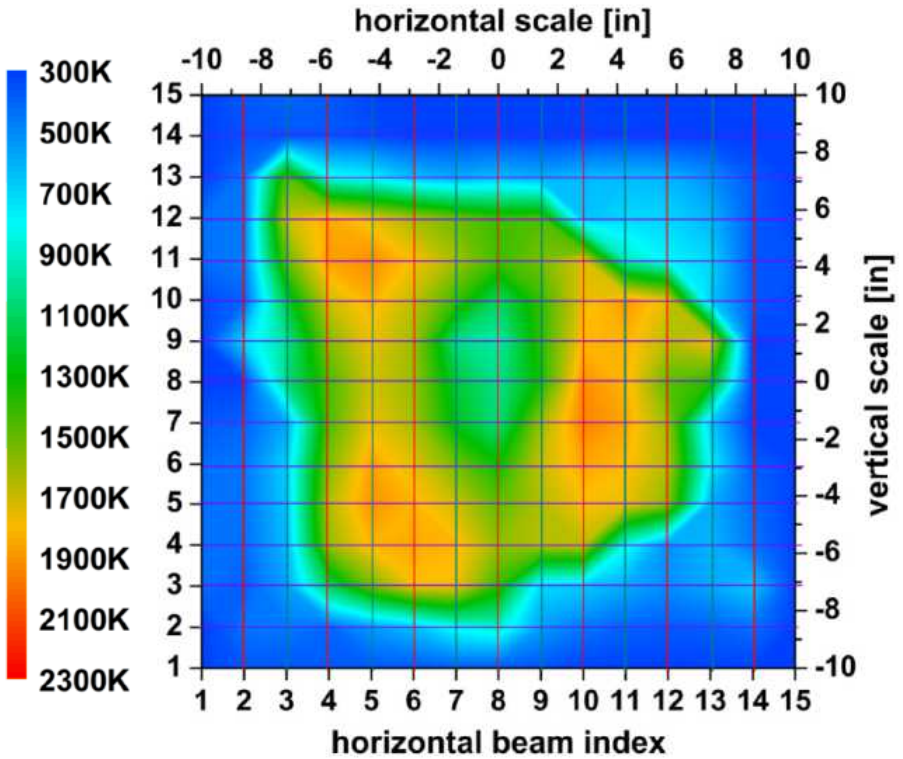


- Measure water and temperature at 50 kHz rate

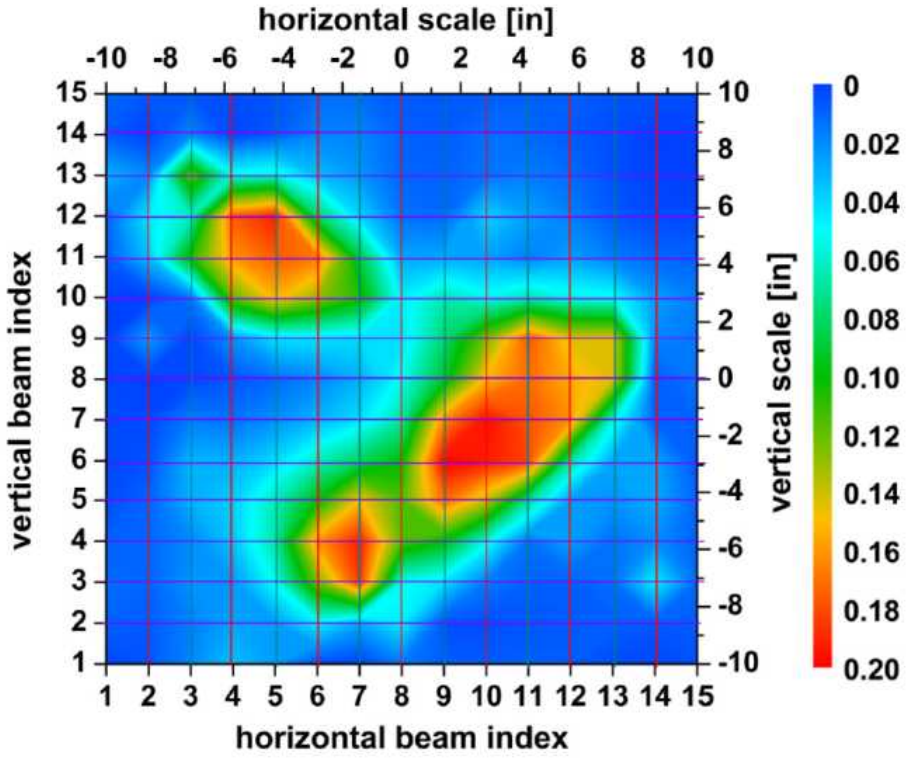
L. Ma, X. Li, S. T. Sanders, A. W. Caswell, S. Roy, D. H. Plemmons, and J. R. Gord, "50-kHz-rate 2D imaging of temperature and H_2O concentration at the exhaust plane of a J85 engine using hyperspectral tomography," Optics Express, Vol. 21, Issue 1, pp. 1152-1162, 2013

TDLAS Tomography Results

Temperature



Water Vapor

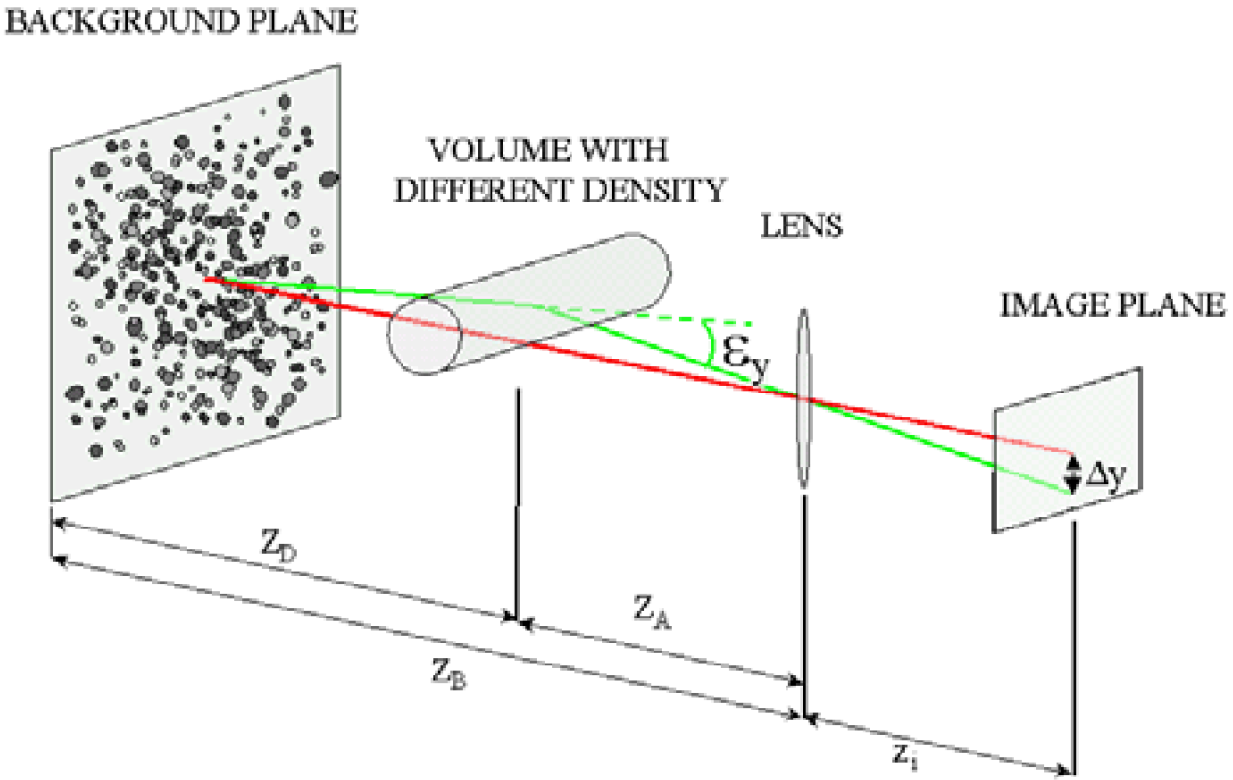


- Measurement is over ~0.5 meter square
  - Time resolution is 20 μsec (50 kHz)
  - Spatial resolution is ~36 mm

L. Ma, X. Li, S. T. Sanders, A. W. Caswell, S. Roy, D. H. Plemmons, and J. R. Gord, "50-kHz-rate 2D imaging of temperature and H_2O concentration at the exhaust plane of a J85 engine using hyperspectral tomography," Optics Express, Vol. 21, Issue 1, pp. 1152-1162, 2013

Background Oriented Schlieren

- BOS is can provide flow visualization
  - Sensitive to density gradients
  - Can measure quantitative density

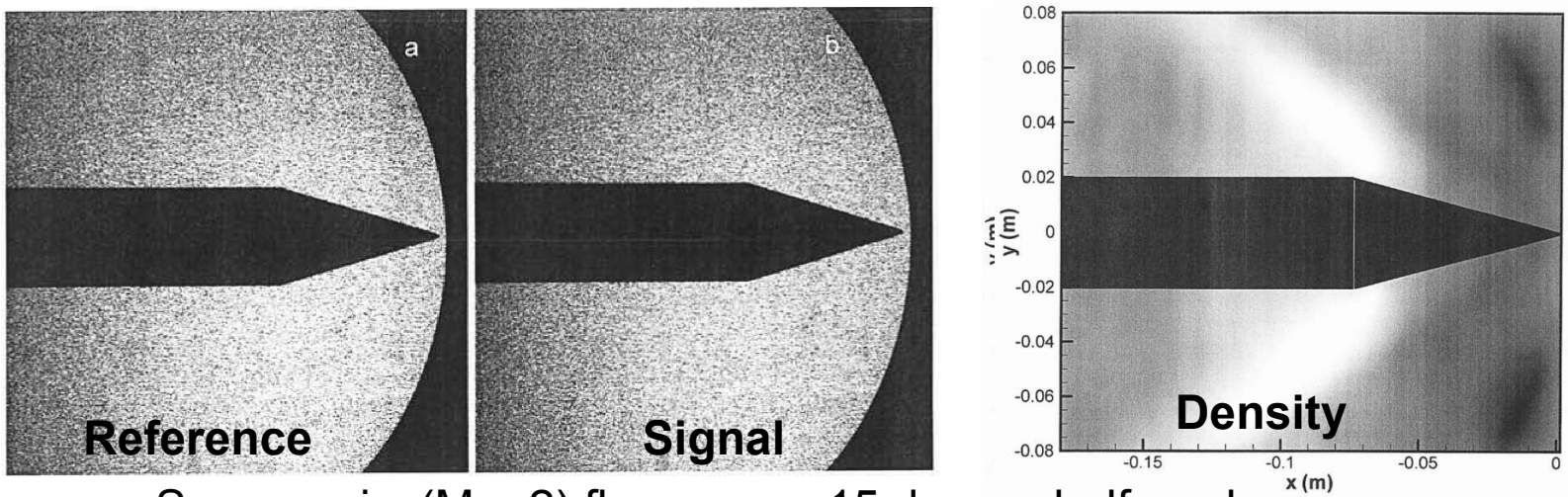


- No laser required!

H. Richard, and M. Raffel. "Principle and applications of the background oriented schlieren (BOS) method." Measurement Science and Technology 12, no. 9, p. 1576-1585, 2001

BOS in supersonic flow

- Acquire reference and signal images
  - Use PIV algorithm to compute displacement



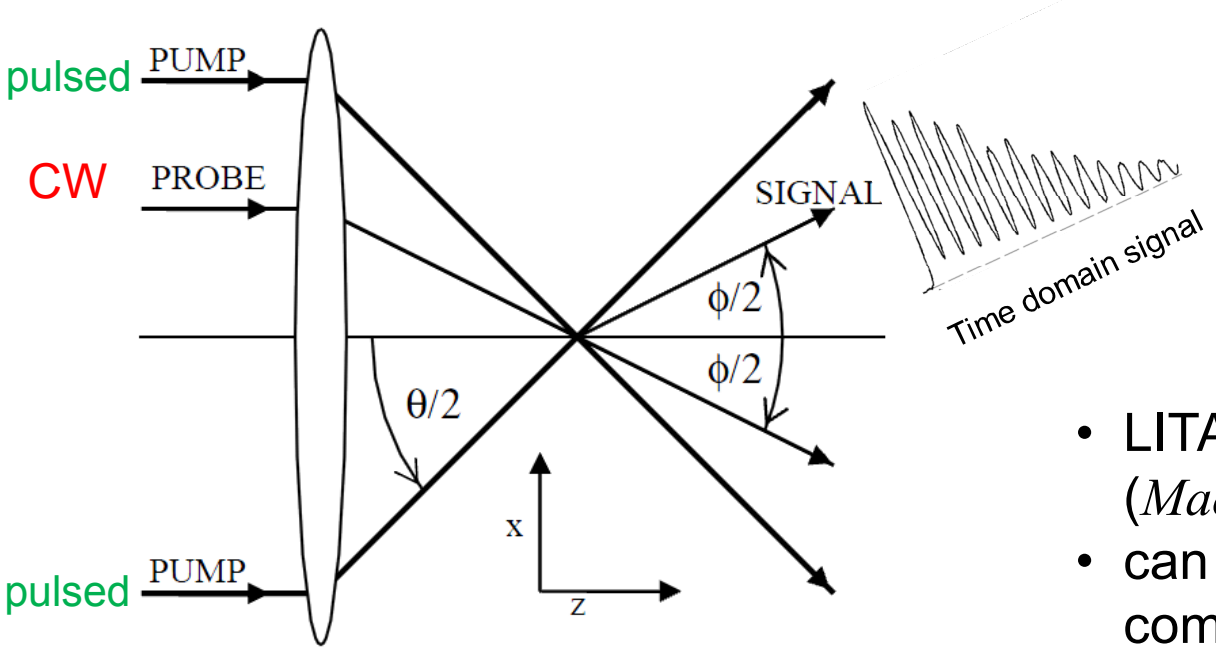
Supersonic (M = 2) flow over a 15 degree half-angle cone

  - Need tomographic deconstruction algorithm for ρ
    - Axisymmetric flow: Abel inversion or other
    - Non-axisymmetric flow:
      - 1 camera, multiple views (rotate model): not time resolved
      - Multiple cameras: can do time-resolved 3D flows

L. Venkatakrishnan, and G. E. A. Meier. "Density measurements using the background oriented schlieren technique." Experiments in Fluids 37, no. 2, p. 237-247, 2004

Laser Induced Thermal Acoustics (LITA)

- Similar to CARS, LITA is a nonlinear measurement technique using 3 input laser beams and generating a 4th beam through a nonlinear interaction with the medium



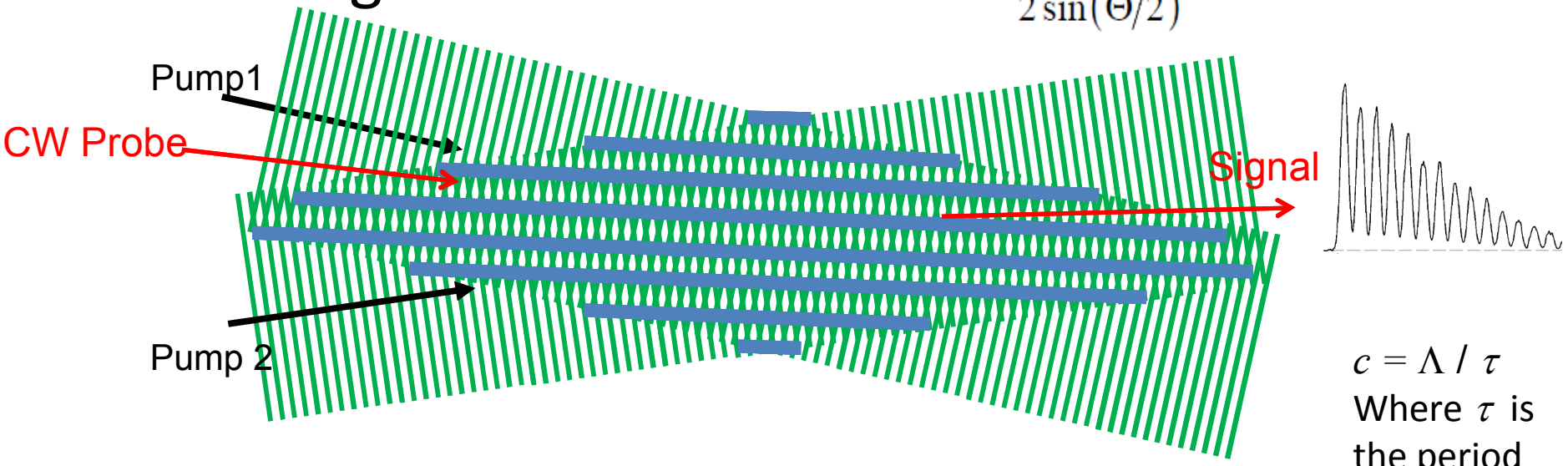
- LITA can measure V, c, $(Mach = V/c)$ and P
- can infer T from c if composition is known

Jenkins, L. N., Hart, R. C., Balla, R. J., Herring, G. C., Khorrami, M. R., & Choudhari, M. M. (2003, November). Application of laser-induced thermal acoustics to a high-lift configuration. In *Optical Science and Technology, SPIE's 48th Annual Meeting* (pp. 134-144). International Society for Optics and Photonics.

Laser-induced gratings

Grating Formation:

$$\text{period} = \Lambda = \frac{\lambda}{2\sin(\Theta/2)}$$



$c = \Lambda / \tau$
Where τ is the period

- Pump 1 and Pump 2 form a grating
  - Thermal grating
  - Electrostriction grating
  - Both result in index of refraction grating

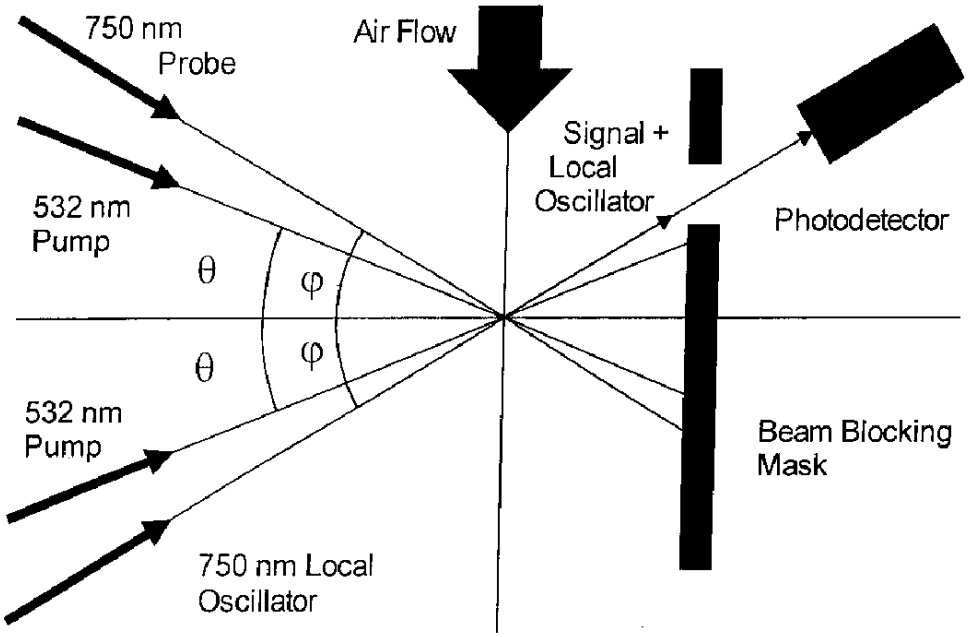
CARS: Spectral domain
LITA: Time domain

Grating Readout

- Probe beam scatters off, reads out the grating
- Signal beam is detected

LITA Measurement Systems

- Use of heterodyne detection increases accuracy.

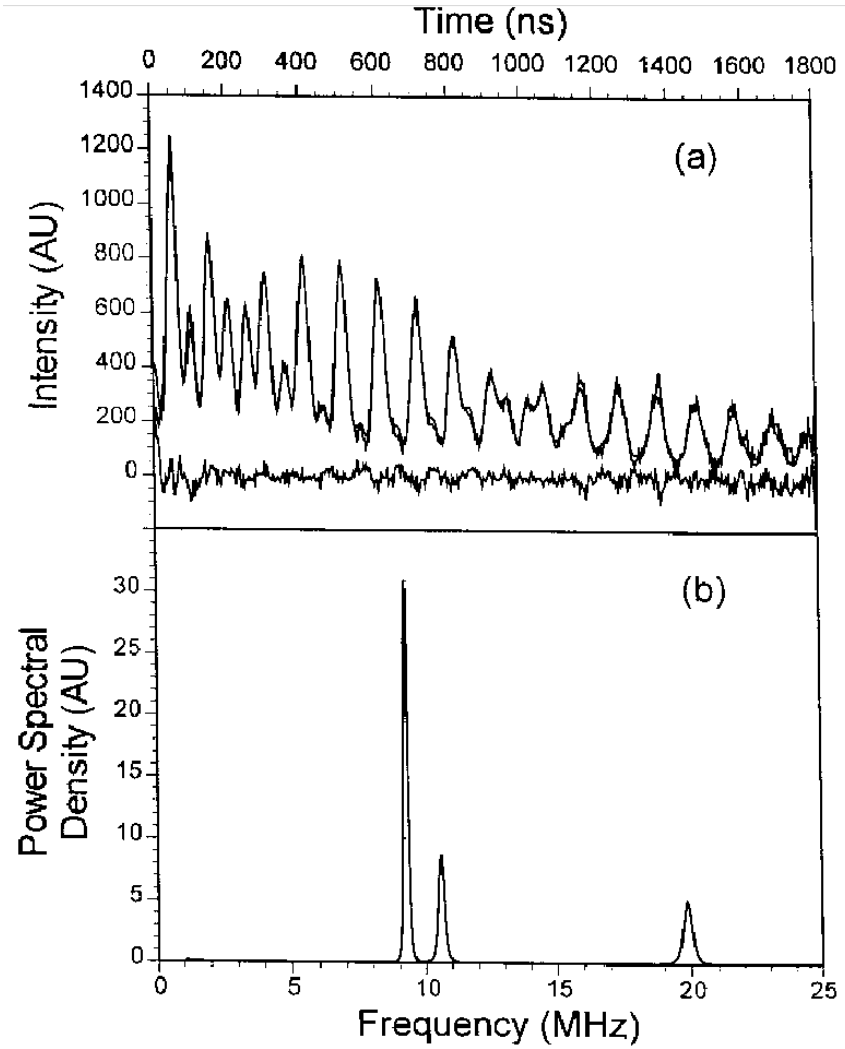


Hart, R. C., Balla, R. J., & Herring, G. C. (2001). Simultaneous velocimetry and thermometry of air by use of nonresonant heterodyned laser-induced thermal acoustics. *Applied Optics*, *40*(6), 965-968.

Schlamp, S., Cummings, E. B., & Sobota, T. H. (2000). Laser-induced thermal-acoustic velocimetry with heterodyne detection. *Optics Letters*, *25*(4), 224-226.

LITA T, V measurement

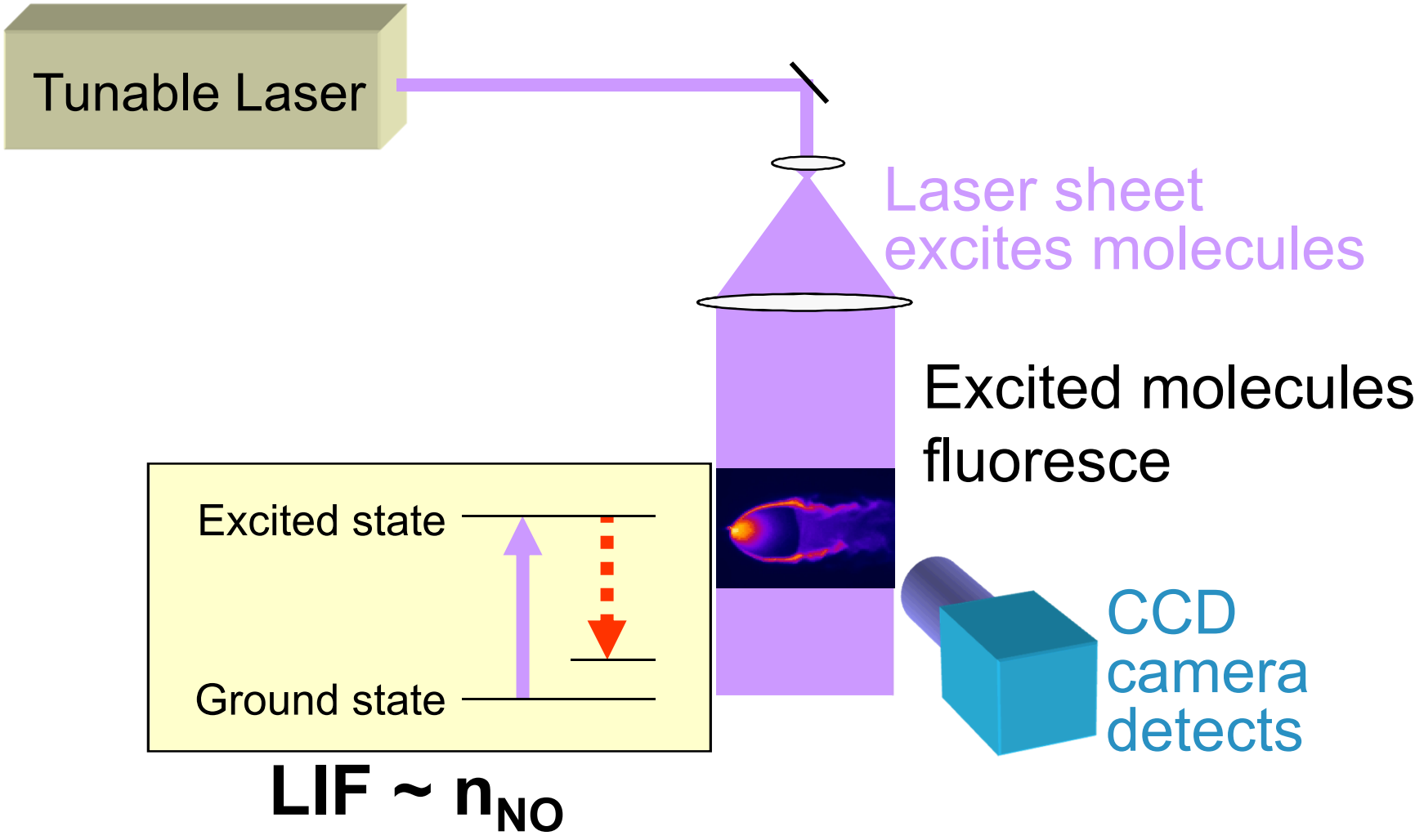
- Time domain signal can be processed to obtain a frequency spectrum showing the sound speed and the velocity
  - Beat frequencies: T, V, M
- Precisions:
  - V ~ 1 m/s (~1% at 150 m/s)
  - T ~ 1 K (~0.4% at 300 K)
- Accuracy
  - V ~ 0.2%
  - T ~ 0.3%
- Probe volume ~1-2 cm
  - Complicates measurement and interpretation in transitional / turbulent flows
  - Nonlinear: Not simply an average over probe volume



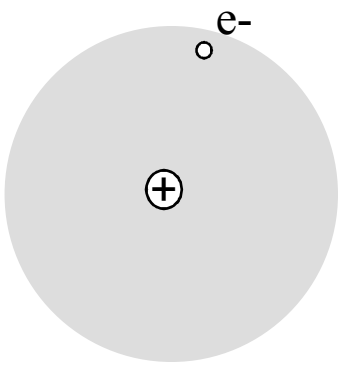
Hart, R. C., Balla, R. J., & Herring, G. C. (2001). Simultaneous velocimetry and thermometry of air by use of nonresonant heterodyned laser-induced thermal acoustics. *Applied Optics*, *40*(6), 965-968.

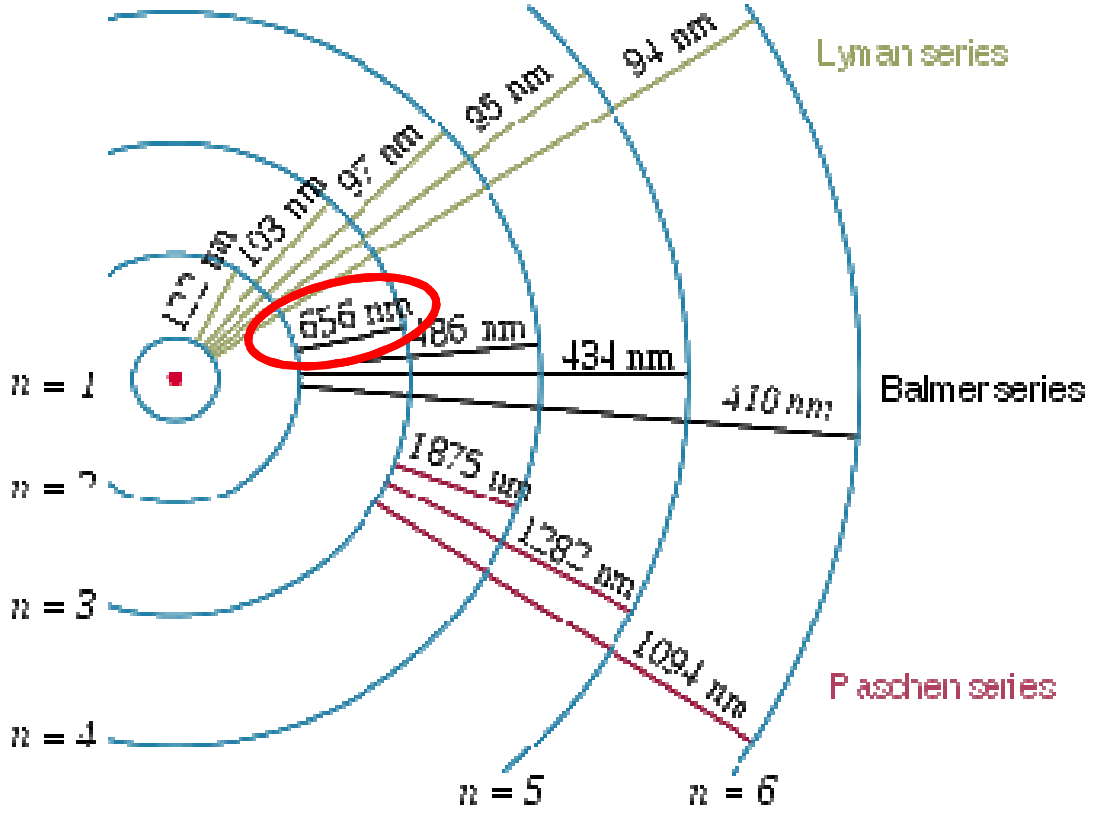
(Planar) Laser-Induced Fluorescence: (P)LIF

Planar Laser-Induced Fluorescence (PLIF)



LIF Theory 1



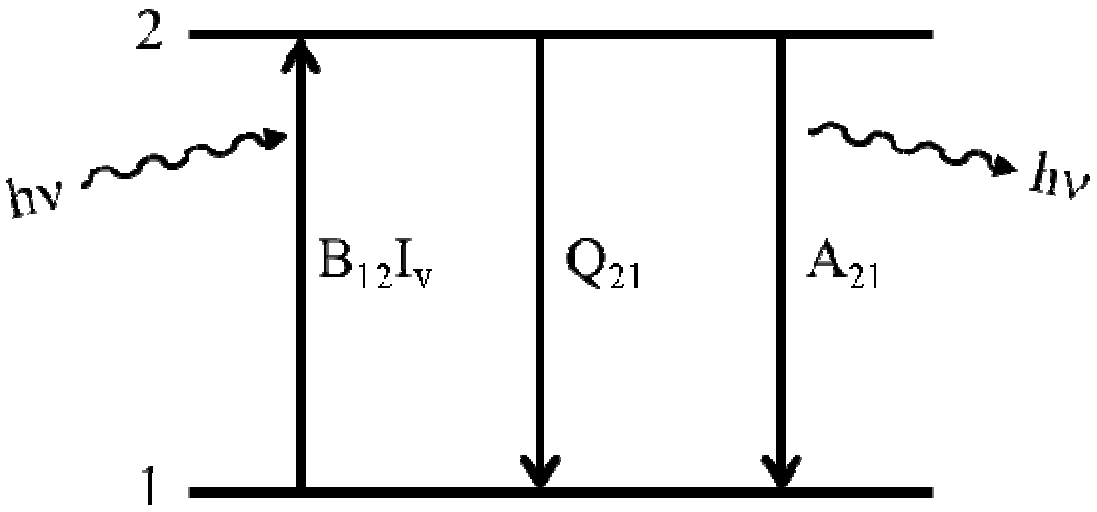


λ= 656 nm
ν= 4.6x10^{14} Hz
$\bar{\nu}$= 15,240 cm^{-1}

- Two level model
  - e.g. Hydrogen atom
- Photon energy = $h\nu$
  - $c = \lambda\nu$ wavenumbers = $\bar{\nu} = 1/\lambda$ cm^{-1}

Hydrogen atom energy levels
(from Wikipedia, "hydrogen spectral series")
http://creativecommons.org/licenses/by-sa/3.0/deed.en

LIF Theory 2



- Two level model
  - Ignore all other levels (rotations, vibrations, etc.)
  - Laser only promotes atoms/molecules from 1 to 2
    - Weak excitation limit, so $N_2 << N_1$
  - Processes:
    - absorption ($B_{12}I_\nu$), quenching (Q_{21}), spontaneous emission (A_{21})

LIF Theory 3

- Absorption, quenching, emission:
  - Probability of an <u>absorption</u> transition per sec = $B_{12}I_\nu = W_{12}$
    - » B_{12} is the Einstein B coefficient
    - » I_ν is the laser's spectral irradiance (W/cm^2/cm^{-1}) at the transition's frequency
    - » $W_{12} = B_{12}I_\nu = B_{12}IG$ (G is the spectral overlap integral)
  - Probability of a <u>quenching</u> collision per sec = Q_{21}

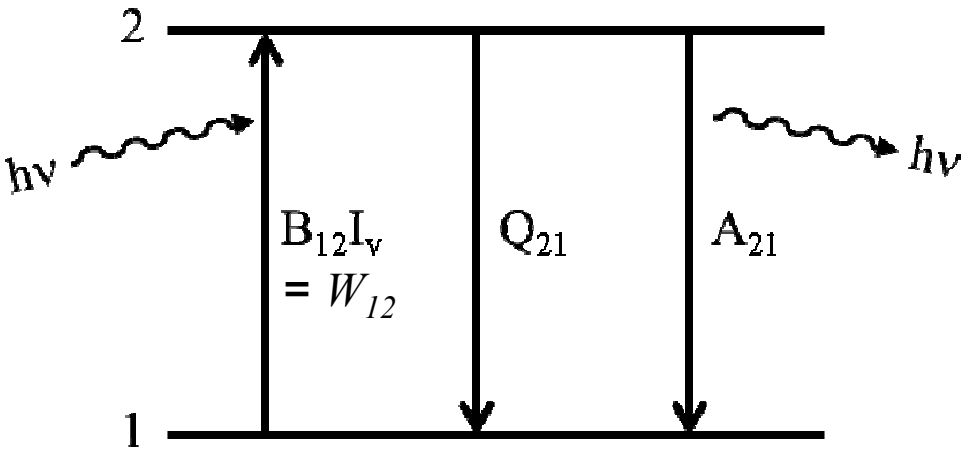


$$Q_{21} = N_T \sum_i \chi_i \sigma_{s,i} v_{s,i}$$

$N_T = P/k_B T$
χ is mole fraction
σ is quenching cross section
v is mean relative velocity

  - Probability of an atom/molecule in excited state undergoing <u>spontaneous emission</u> per second = A_{21}
    - » Einstein A coefficient

LIF Theory 4



- Rate equation analysis:

$$\frac{dN_1}{dt} = -N_1 W_{12} + N_2(Q_{21} + A_{21}) = -\frac{dN_2}{dt}$$

$$N_1 + N_2 = N_S = \chi_s f_B N_T \qquad N_T = P/k_B T$$

Number density of the species starting in that state before excitation

Boltzmann Fraction, f_B fraction of molecules in the state being probed

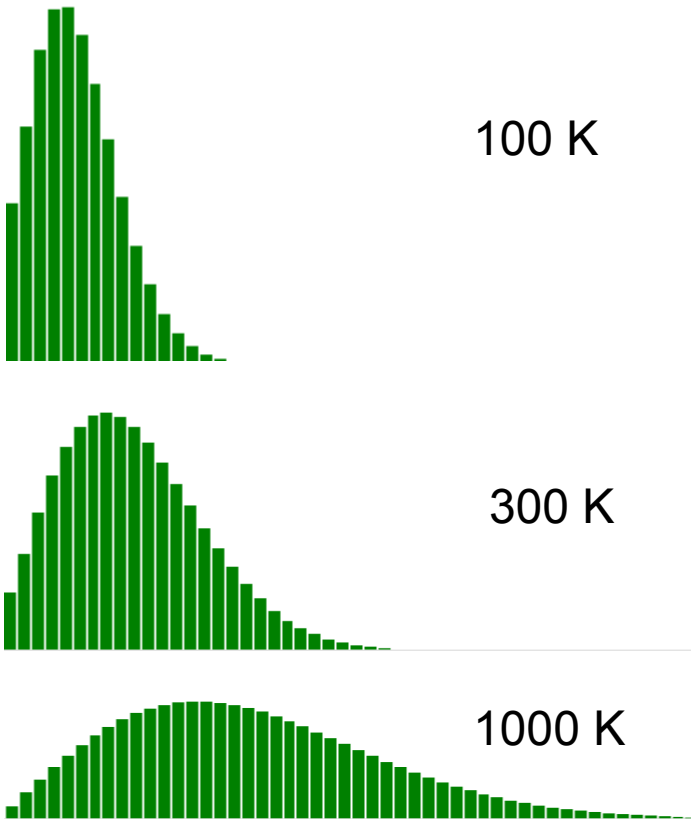
$$f_B = \frac{g_j e^{-E_j/k_B T}}{\sum_j g_j e^{-E_j/k_B T}}$$

Boltzmann Fraction

- Rotational Boltzmann Distributions

$$f_B = \frac{g_j e^{-E_j/k_B T}}{\sum_j g_j e^{-E_j/k_B T}}$$

  - a function of temperature only
  - Peak at 300 K is ~3% for $N = 7$ ($J = 7.5$)
- Vibrational and electronic Boltzmann fractions
  - Most molecules in ground state for $T<2000$ for atoms, diatomics



Computed using LIFBASE
(J. Luque, SRI International)

LIF Theory 5

- Steady state solution:
  - cw laser, or many collisions during laser pulse
  - Time derivatives → 0
  - 2 algebraic equations. Solve for N_2:

$$N_2 = \frac{\chi_s f_B N_T W_{12}}{W_{12}+Q_{21}+A_{21}}$$

  - Emission *rate* = $N_2 A_{21}$

$$S_{LIF} = \chi_s f_B N_T B_{12} I G \Phi t_{det} V \frac{\Omega}{4\pi} \eta$$

total number of photons collected

detection time

volume excited

solid angle

collection efficiency

  - Where we define the fluorescence yield:

$$\Phi = A_{21}/(Q_{21}+A_{21})$$

Weak excitation:
$W_{12} << Q_{21}+A_{21}$

LIF Theory 6

- Steady state solution: dependencies

$$S_{LIF} \propto \chi_s f_B(T) B_{12} IG(\chi_s, P, T, U) \Phi(\chi_s, \chi_i, P, T) t_{det}$$

  - LIF signal is sensitive to T, P, U, χ_s, χ_i
    - Can potentially measure these parameters
    - Complicates measurement of a single parameter
  - Making quantitative measurements with LIF involves finding a way to make measurement sensitive only to the parameter being measured

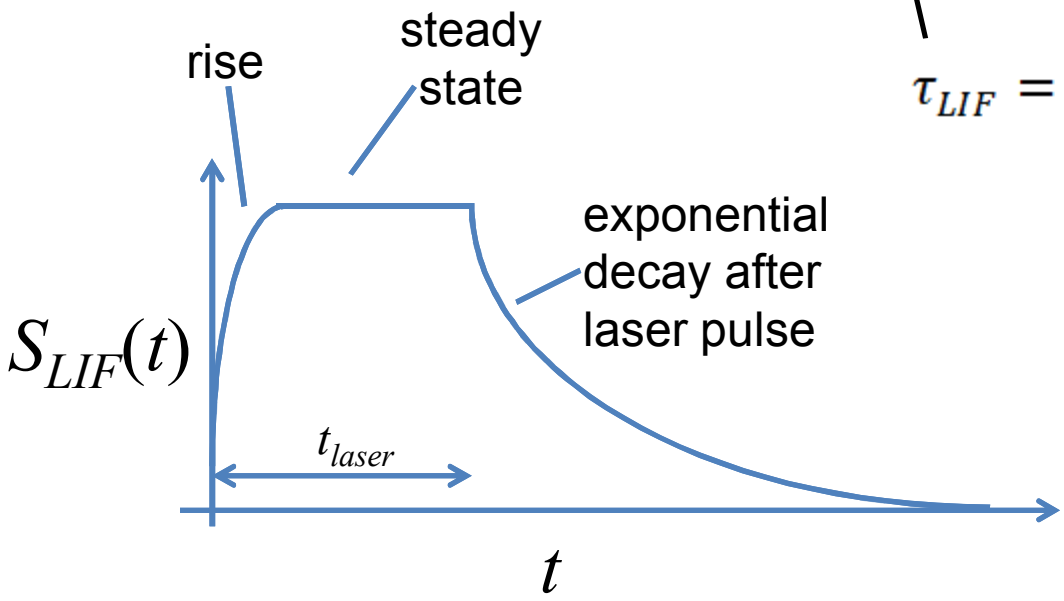
LIF Theory 7

- Time dependent solution:

$$S_{LIF}(t) = S_{LIF}(t_{laser})e^{-(t-t_{laser})/\tau_{LIF}},\ t > t_{laser}$$

photons collected per time

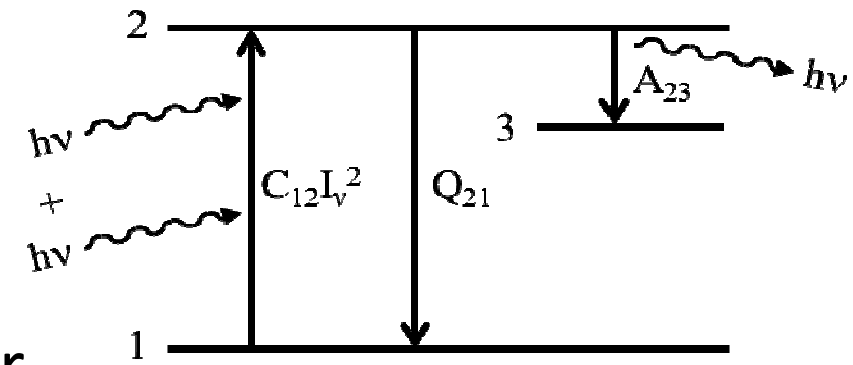
$$\tau_{LIF} = (Q_{21} + A_{21})^{-1}$$



  - Laser pulse length is typically ~10 ns
  - LIF lifetimes can be a few ns to 100's of ns or longer
    - If you can measure lifetime, you can measure Q!
    - Can measure velocity from long lived decay (MTV)

LIF Theory 8

- More complete LIF theories and phenomena:
  - Saturation effects (see manuscript)
  - Line shapes
    - overlap integral, G, to be discussed below
  - Multi-level LIF models
    - 2 photon absorption
    - 3, 4, 5 level models
  - Predissociation
  - Rotational energy transfer
  - Vibrational energy transfer



2-photon LIF, 3 level model

- For further reference:

A. C. Eckbreth, *Laser Diagnostics for Combustion Temperature and Species 2nd Ed.*, Combustion Science & Technology Book Series, Volume 3, Taylor & Francis, New York, 1996

A real molecule: NO

- NO is a diatomic molecule
  - Electronic
  - Vibration
  - Rotation
    - Not shown
- Absorption and fluorescence take place between electronic states
  - Show rotational and vibrational structure

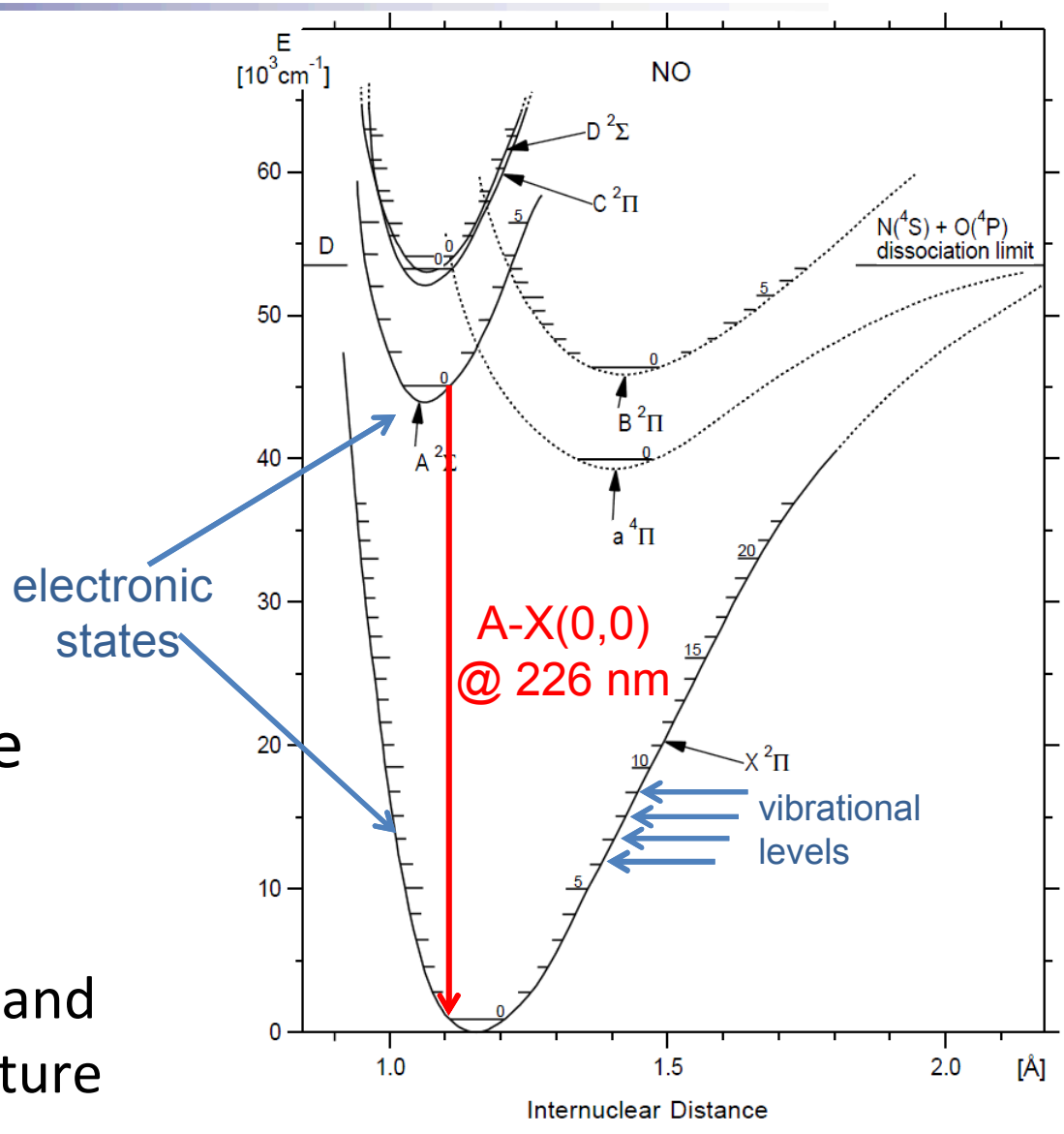
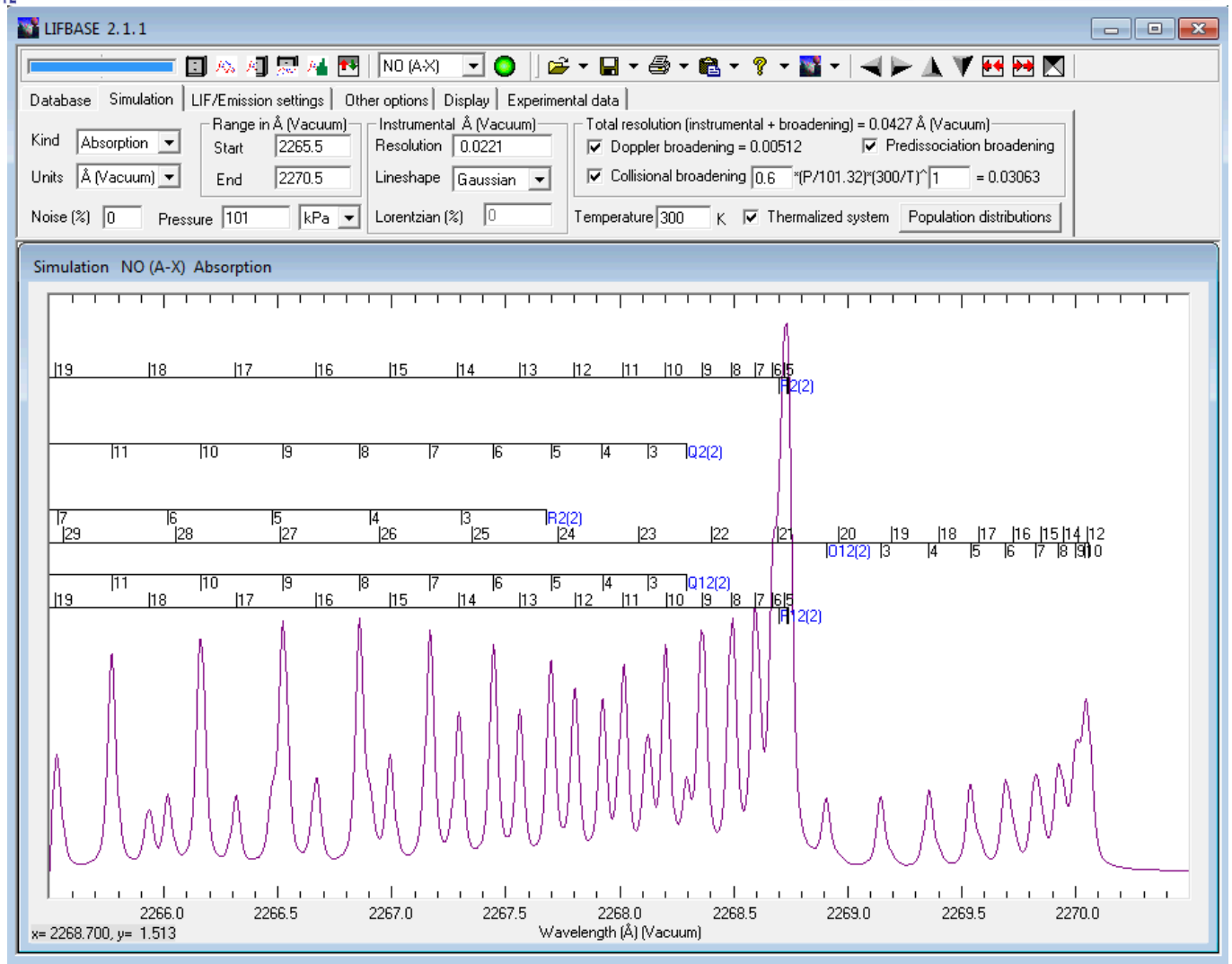


Figure A2.1. Selected potential energy curves that show the electronic structure of NO. In this experiment, we probed transitions in the A–X band of NO [adapted from (Scheingraber and Vidal,1985

LIFBASE Simulation of NO



LIFBASE freeware by J. Luque, SRI International

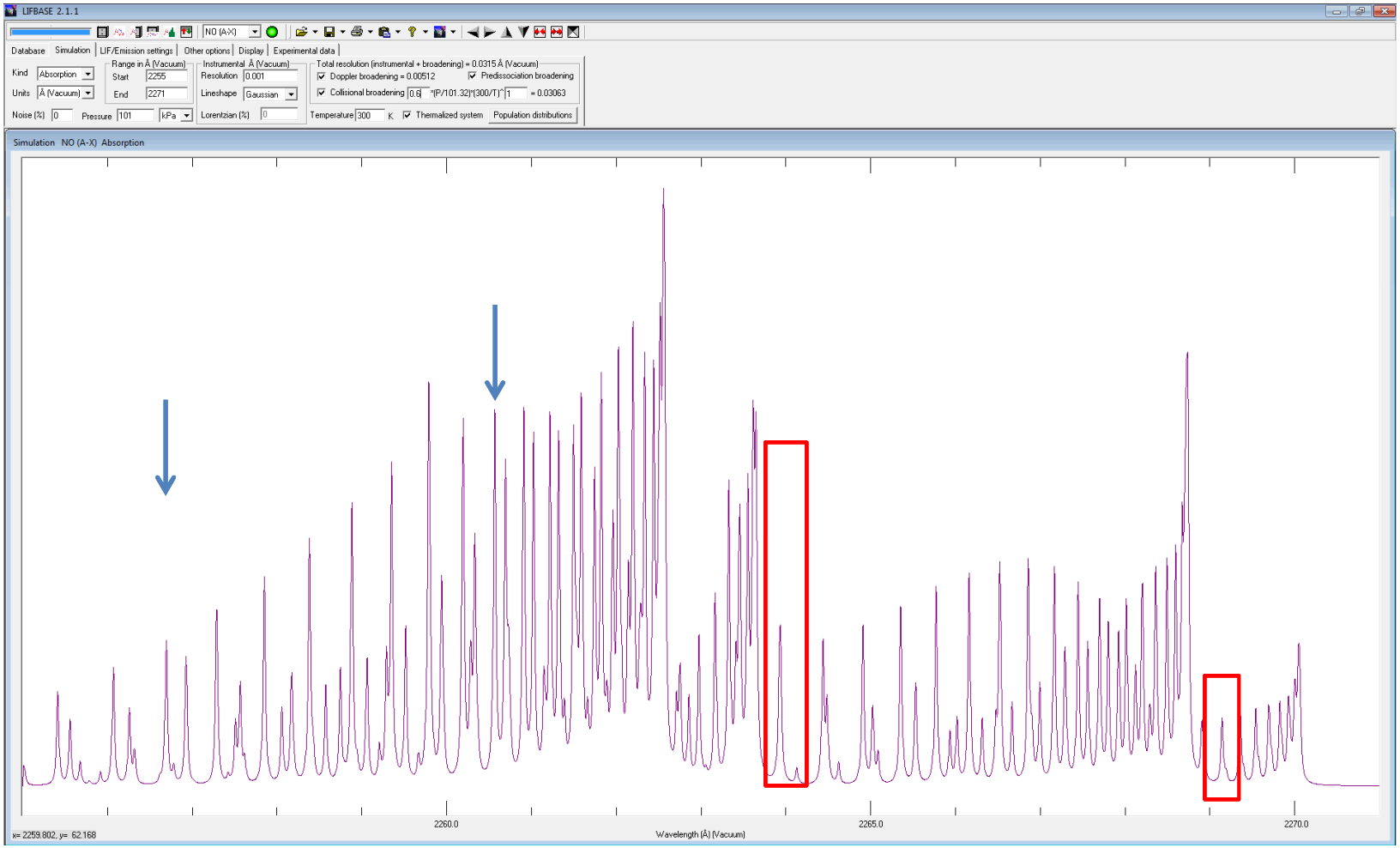
lifbase@yahoo.com

- LIF Excitation Scan between 226.6-227 nm, 1 atm, 300 K
  - Lines all have band names (*O, P, Q, R, S*) and *N*'s or *J*'s (rot quant #)

LIF Temperature Measurements

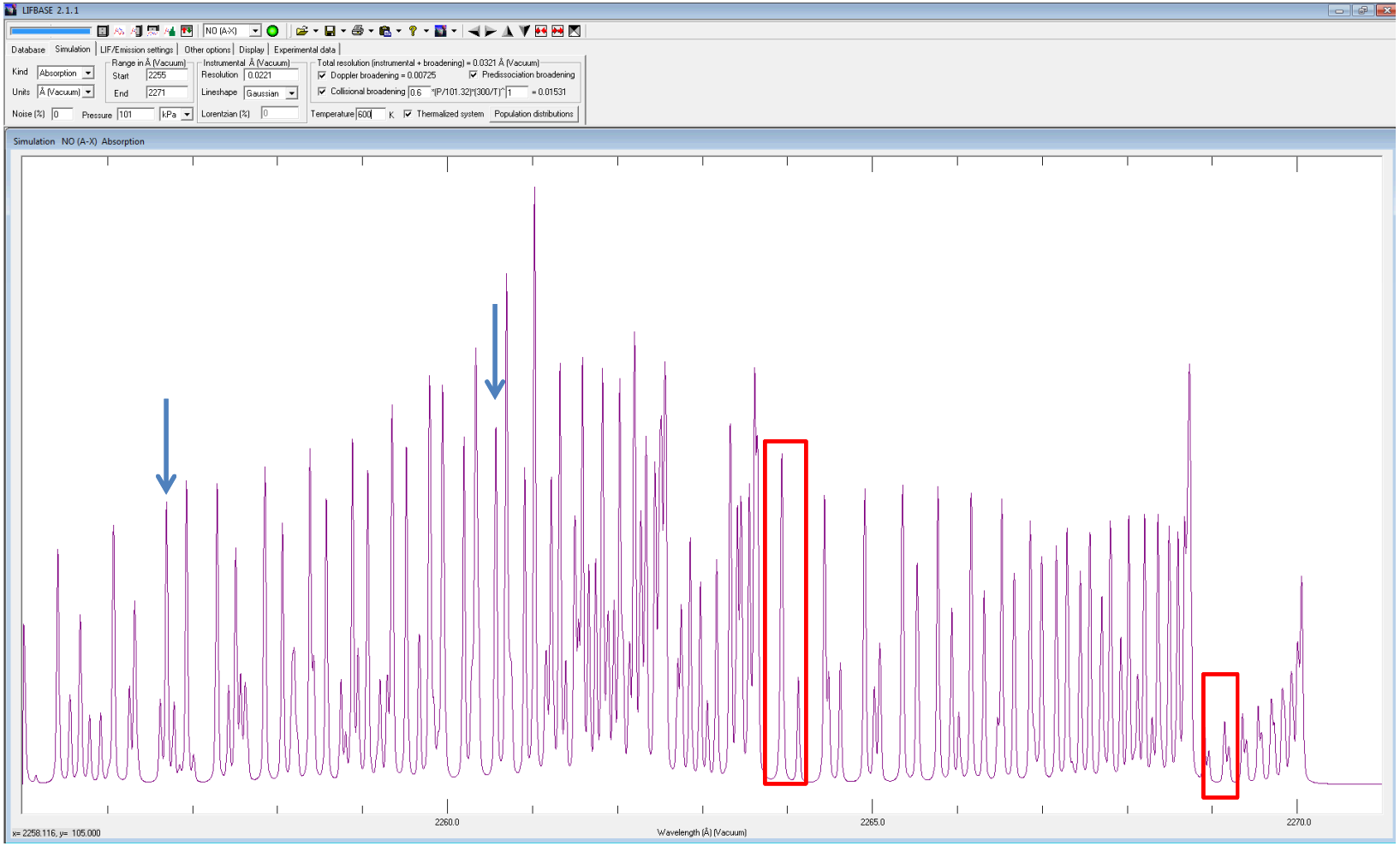
- Rotational Temperature
  - Ratio of 2 or more rotational lines
  - Describe general case
  - Example in a turbulent base flow
- Vibrational Temperature
  - Ratio of 2 or more vibrational lines
  - Example of freestream characterization
    - Boltzmann plot, where more than 2 lines used
- Translational Temperature

Rotational Lines of NO, 300 K



- LIF Excitation Scan between 225-227 nm, 1 atm, 300 K

Rotational Lines of NO, 600 K



- LIF Excitation Scan between 225-227 nm, 1 atm, 600 K

LIF: Rotational Temperature, T_{rot}

- Recall:

$$S_{LIF} \propto \chi_s f_B(T) B_{12} I G(\chi_s, P, T, U) \Phi(\chi_s, \chi_i, P, T) t_{det}$$

- Obtain LIF or PLIF on two different rotational lines of a molecule: i and j. Take ratio, R:

$$\frac{S_{LIF,i}}{S_{LIF,j}} = R = C \frac{B_{12,i} E_i}{B_{12,j} E_j} \frac{(2J_i+1)\exp[-F_{J,i}/k_B T_{rot}]}{(2J_j+1)\exp[-F_{J,j}/k_B T_{rot}]}$$

- Where:
  - C is a constant
  - E is the laser energy (in mJoules)
  - J is the rotational quantum number
  - F_J is the energy of the state
  - k_B is Boltzmann's constant

Ω, χ_s, η, G, V, N_T and Φ (including Q_{21}) *cancel out!*

LIF: Rotational Temperature, T_{rot}

- Solve for T_{rot}:

$$T_{rot} = \frac{-\Delta E_{rot}}{k_B \ln\left[CR\frac{B_{12,j}E_j(2J_j+1)}{B_{12,i}E_i(2J_i+1)}\right]}$$

- Where:

$$\Delta E_{rot} = (F_{J,i} - F_{J,j})$$

- Temperature sensitivity:

$$\frac{\delta T_{rot}}{T_{rot}} = \frac{k_B T_{rot}}{\Delta E_{rot}} \frac{\delta R}{R}$$

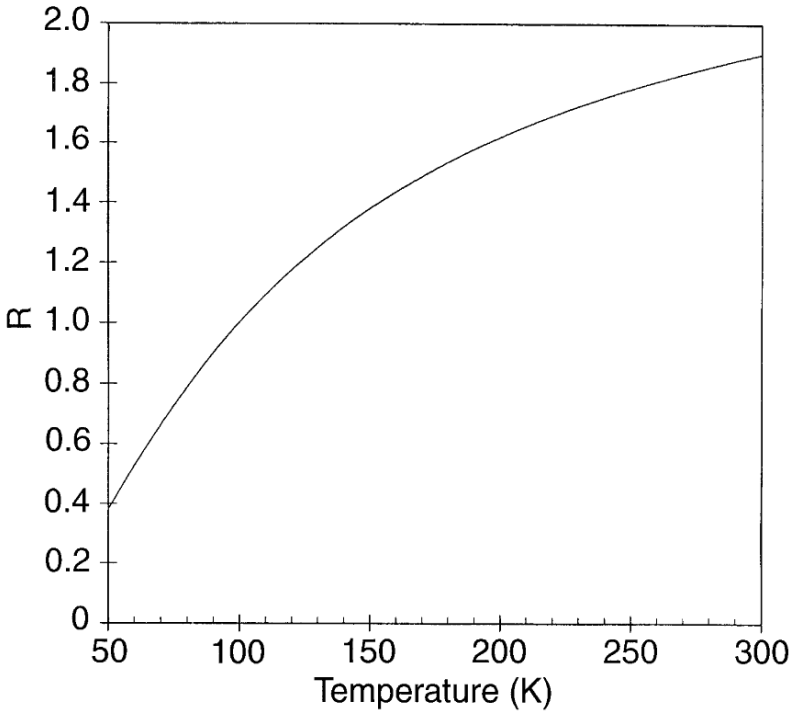
- Suggests to maximize ΔE_{rot} (as long as signals still strong) to minimize error in temperature, δT_{rot}
  - Should choose widely separated states: e.g. J = 3 & 35

Typical Strategies for T_{rot}

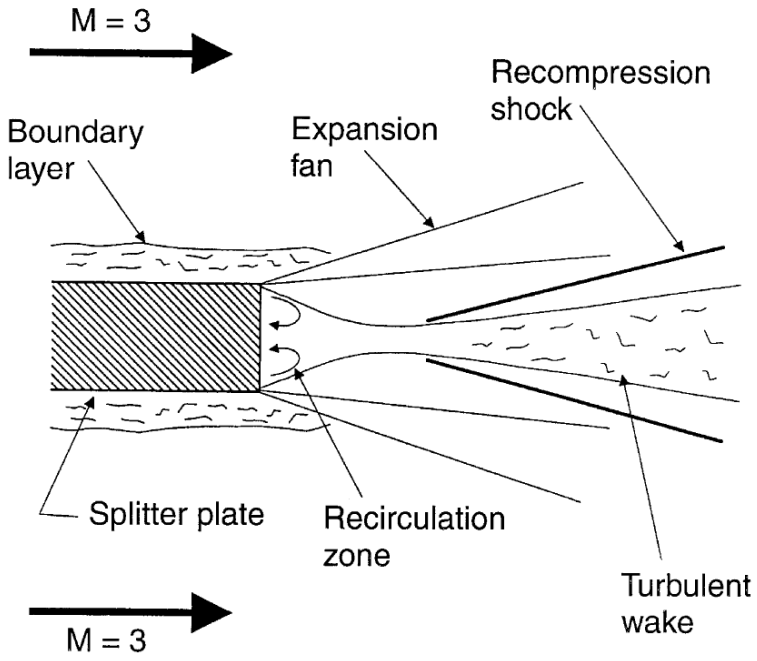
- Typically two approaches:
  - Steady laminar flow
    - Obtain images on one J, then scan laser to another J
    - Ratio images, determine temperature
  - Turbulent flow
    - Two lasers, two cameras (Cumbersome! Expensive!)
    - Acquire two images nearly simultaneously, different J's
    - Need to do this because ratio is a nonlinear function of temperature: $\frac{S_{LIF,i}}{S_{LIF,j}} = R = C\frac{B_{12,i}E_i}{B_{12,j}E_j}\frac{(2J_i+1)\exp[-F_{J,i}/k_BT_{rot}]}{(2J_j+1)\exp[-F_{J,j}/k_BT_{rot}]}$
    - Typical single-shot temperature measurement precision is ~20% at flame temperatures (Seitzman, 1994)
    - If you use 1 laser/1 camera, →bias errors in turb. Flows
      - (E. R. Lachney, and N. T. Clemens, "*PLIF imaging of mean temperature and pressure in a supersonic bluff wake*," Experiments in Fluids, 24, pp. 354-363, 1998.)
      - Average many measurements to reduce uncertainty

Single laser/camera for T_{rot} in turbulent flow

- Lachney and Clemens (1998) used 1 laser / 1 camera to map mean T_{rot} in a base flow.
- NO seeded N_2 at Mach 3
- Chose: $J = 8.5$ and $J = 10.5$



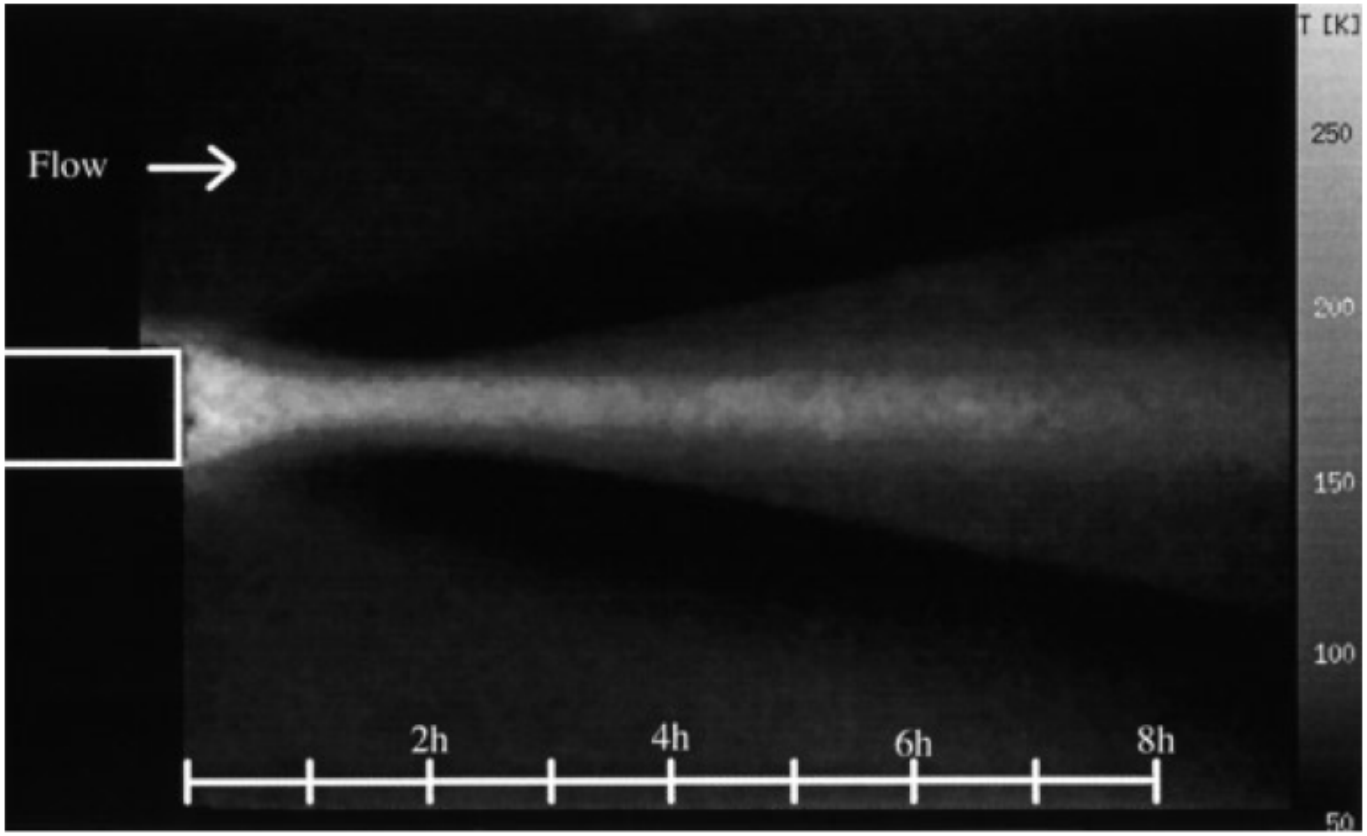
Ratio of lines, used to measure T_{rot}



Turbulent supersonic wake flow

Single laser/camera for T_{rot} in turbulent flow

- Lachney and Clemens (1998):

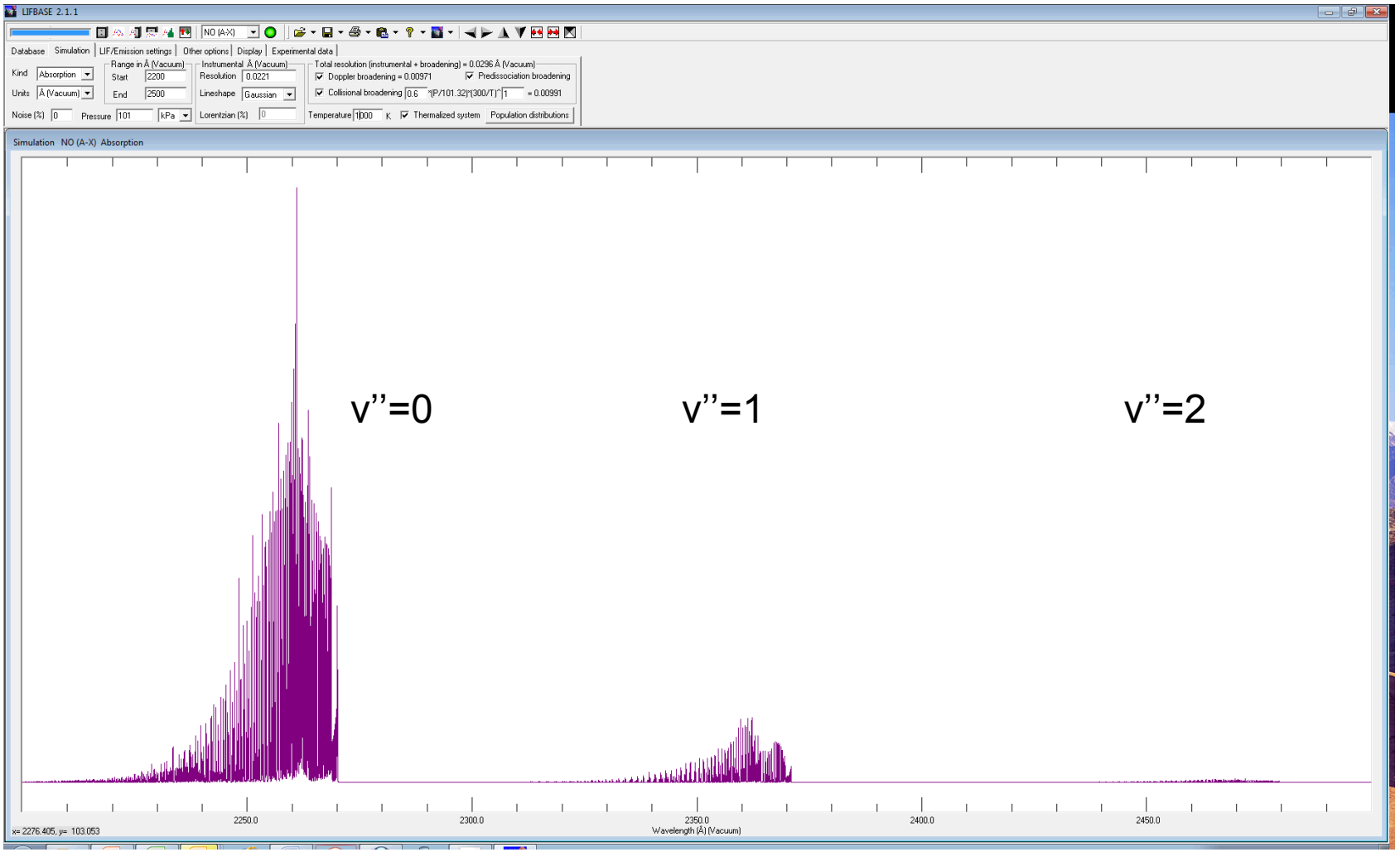


Turbulent supersonic wake flow temperature measurement

Agreed with temperatures inferred from probe measurements to within 11%

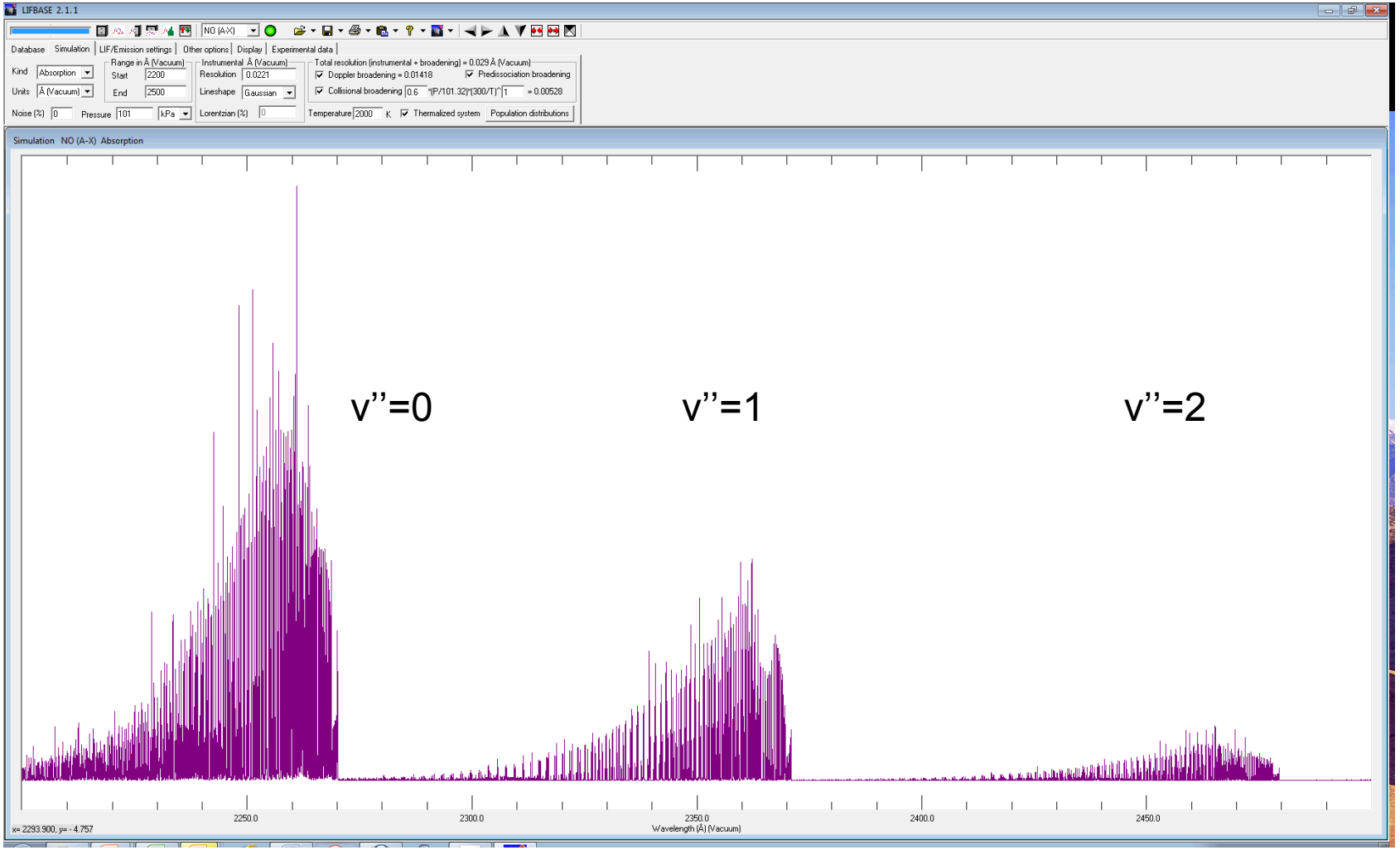
E. R. Lachney, and N. T. Clemens, "*PLIF imaging of mean temperature and pressure in a supersonic bluff wake*," Experiments in Fluids, 24, pp. 354-363, 1998

Vibrational Bands of NO



- Three vibrational bands: T = 1000 K
  - Vibrational bands have quantum number, *v*

Vibrational Bands of NO



- Three vibrational bands: T = 2000 K

Vibrational Temperature T_{vib} Measurement

- A similar approach can be made to obtain vibrational temperature measurements
  - Typically excite the same rotational line (J) in 2 or more vibrational bands ($F_j \approx$ constant)
  - Can do 2 line measurements as before
  - Can use a Boltzmann Plot:
    - Graph:

$$\ln\left[\frac{S_{LIF,i}}{B_{12,i}E_i(2J_i+1)\exp[-F_{J,i}/k_BT_{rot}]}\right]$$

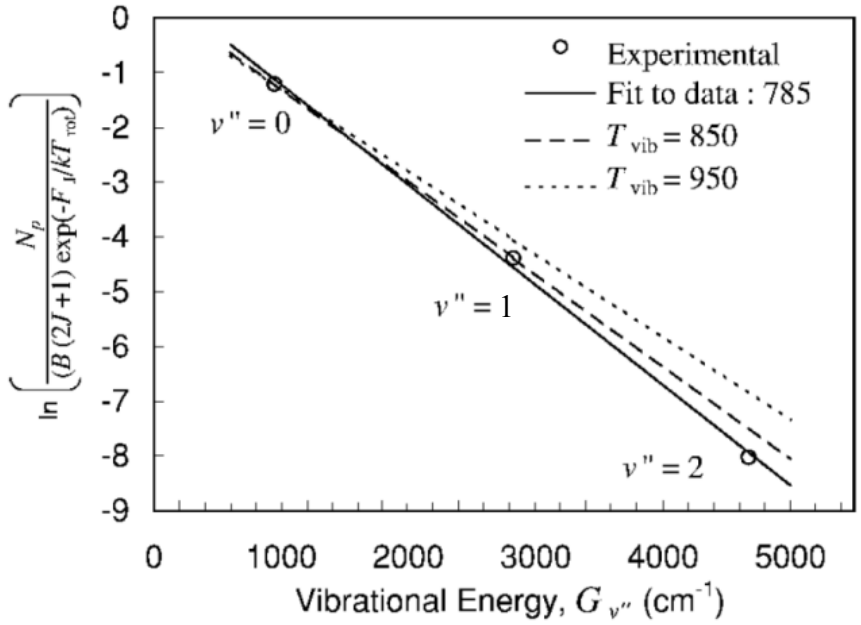
versus $G_{v,i}$ (vibrational energy of states probed)

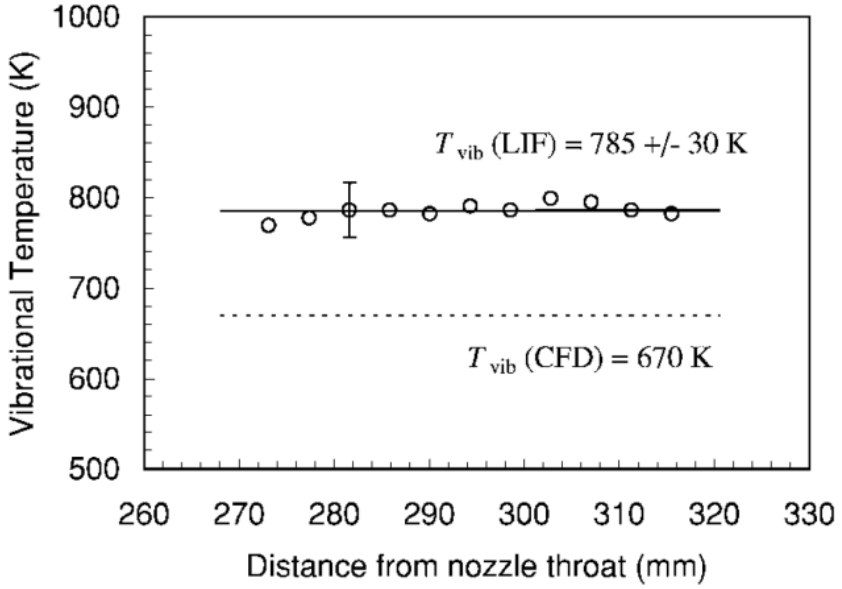
    - Slope is: $-1/(k_BT_{vib})$

If $F_j \approx$ constant then T_{vib} can be determined independent of T_{rot}

Vibrational Temperature T_{vib} Measurement

- Vibrational temperature measured in a nozzle exit flow verifying vibrational nonequlibrium



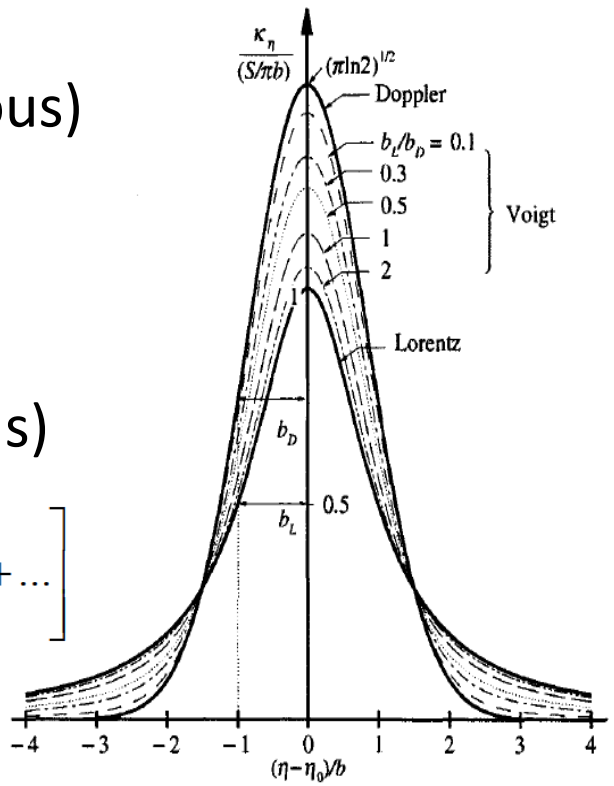


$T_{rot} \approx 400$ K and decreasing with distance

P. C. Palma, P. M. Danehy, and A. F. P. Houwing, "*Fluorescence Imaging of Rotational and Vibrational Temperature in Shock-Tunnel Nozzle Flow*," AIAA Journal, 41(9), September 2003

Translational Temperature

- Can measure T_{trans} (and P) from width and shapes of spectral lines
- Spectral lines have a shape
  - Molecular motion leads to:
    - Doppler Broadening (inhomogeneous)
    - Gaussian shape
    - FWHM: $\Delta\nu_D = \frac{\nu_0}{c}\sqrt{\frac{8\ln(2)k_BT}{m}}$
  - Collisions lead to:
    - Pressure Broadening (homogeneous)
    - Lorentzian shape
    - FWHM: $\Delta\nu_P = \frac{P}{101{,}325}\cdot\left[2\gamma\cdot\chi_i\cdot\left[\frac{295}{T}\right]^{0.75}+\ldots\right]$
  - Combination is: Voigt
    - Convolution (if $\Delta\nu_P, \Delta\nu_D$ comparable)
    - Formulas in manuscript



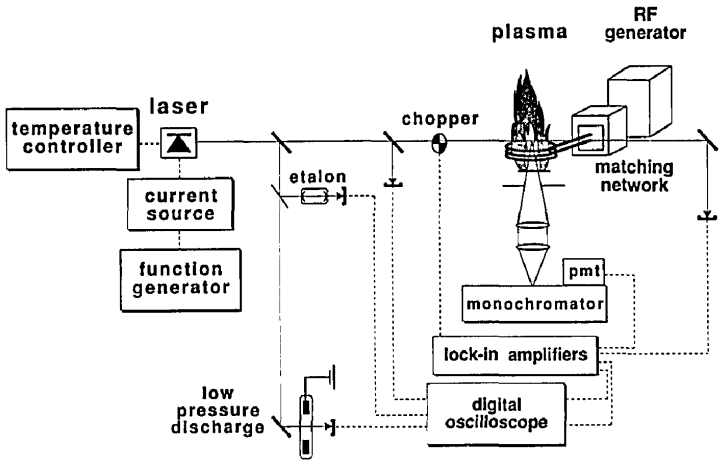
Modest, M: *Radiation Heat Transfer*, Academic Press, Elsevier Science, USA, 2003

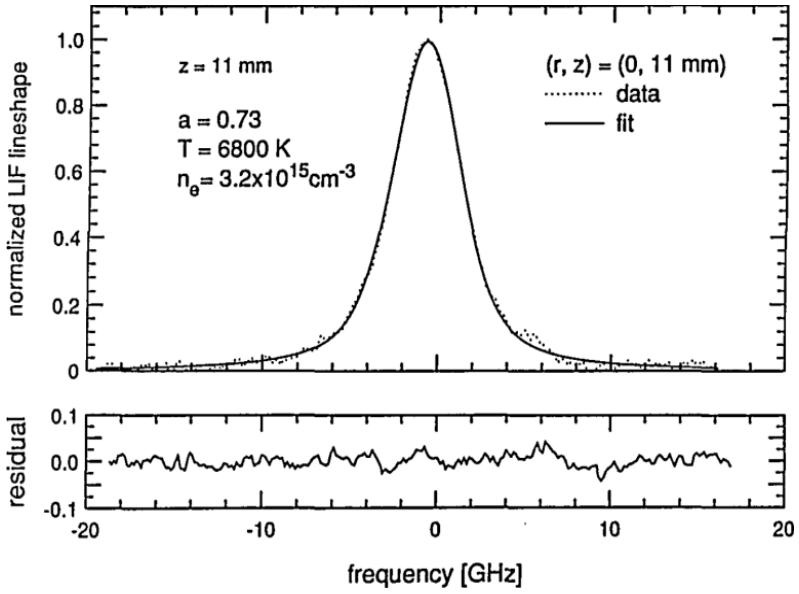
Translational Temperature

- If using a very narrow linewidth laser…

$$\Delta\nu_L << \Delta\nu_P, \Delta\nu_D$$

  - …can neglect laser width
  - Semiconductor diode laser
- Scan laser frequency across a transition
  - Argon atom at 810.4 nm
  - Turbulent inductively coupled plasma (ICP) torch
- Fit measured LIF spectrum
  - Have homogeneous (Stark) and Doppler broadening
  - Determine n_e and T





D. S. Baer, H. A. Chang, and R. K. Hanson, "*Fluorescence diagnostics for atmospheric-pressure plasmas using semiconductor lasers*," Journal of the Optical Society of America B, 9(11), pp. 1968-1978, November, 1992

LIF Velocity

- Doppler shift
  - Based on laser frequency relative to absorption line
- Molecular tagging velocimetry (MTV)
  - aka Flow tagging velocimetry
  - Time of flight
  - Requires lifetime long enough for flow to move while light is being emitted

Doppler Shift Velocimetry

- The Doppler shift:



$$\Delta\nu = \frac{U}{c}\nu_0$$

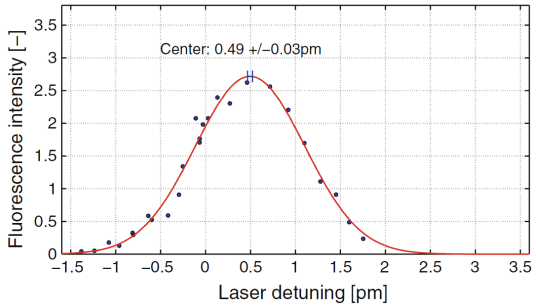
Wikipedia "Doppler Shift"
http://creativecommons.org/licenses/by-sa/3.0/deed.en

- Can be used to measure velocity with LIF
  - Relative shift between laser and absorbing molecules
  - Angle of detection does not matter for LIF
- The overlap integral, G, which describes the overlap of the laser's spectrum with a line
  - L_ν is laser's spectral shape, typically modeled as Gaussian or Lorentzian
  - Y_ν is the transition's absorption line shape, can be shifted by Doppler shift

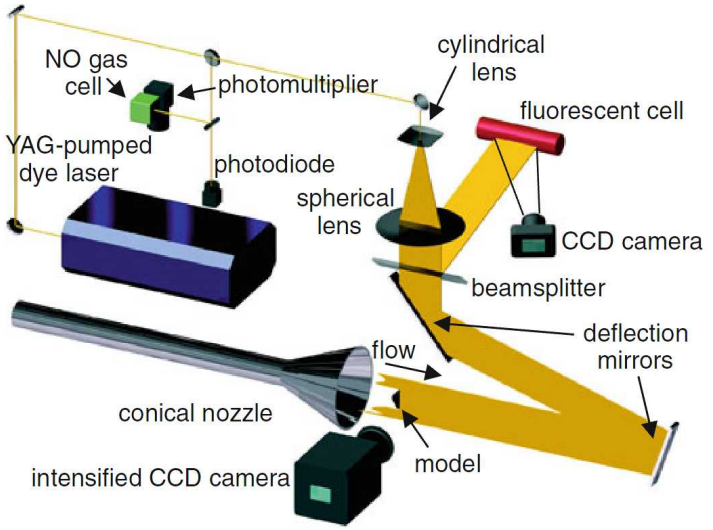
$$G = \int L_\nu Y_\nu d\nu$$

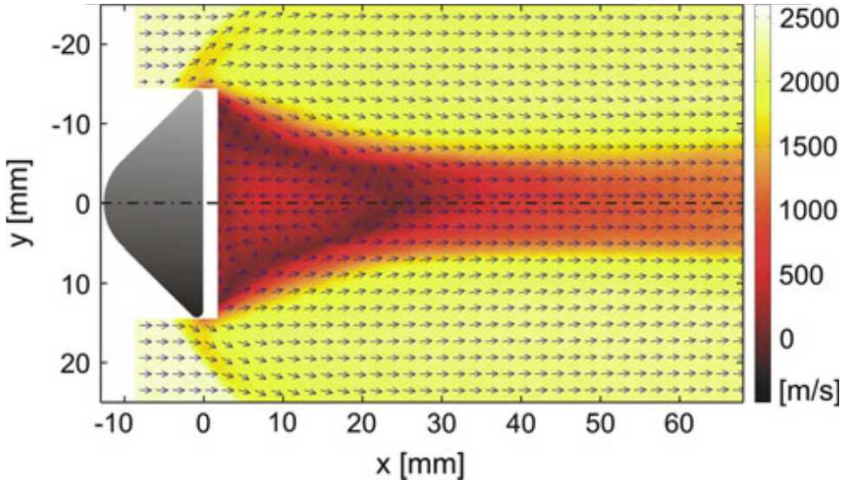
Doppler-based LIF Velocimetry

- Direct laser sheet into flow
- Acquire images with different laser frequencies
  - Measure spectrum at each point



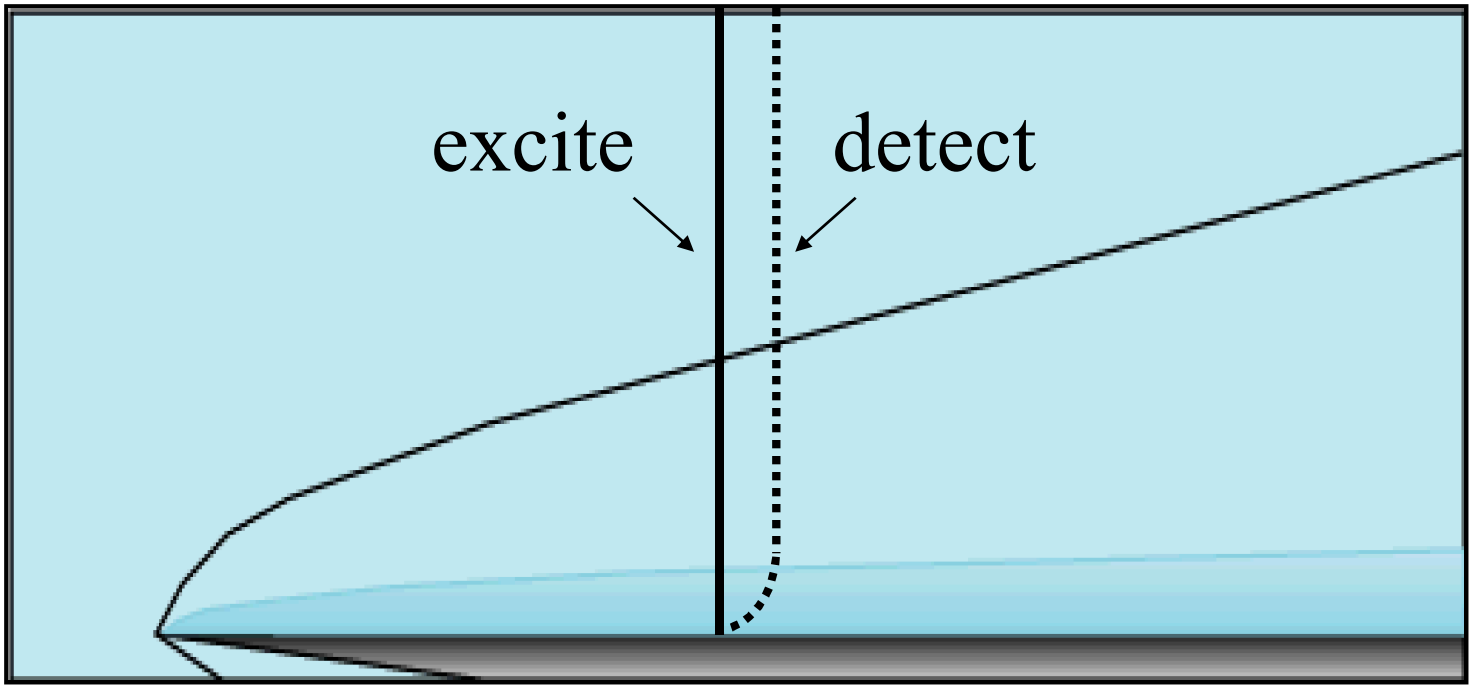
  - Measure from two different directions to get two different velocity components
  - Do need to consider pressure shift
- Fixed frequency (single-shot) schemes also exist but less accurate (see manuscript)





R. Hruschka, S. O'Byrne, and H. Kleine, "*Two-component Doppler-shift fluorescence velocimetry applied to a generic planetary entry probe model*," Experiments in Fluids, 48, pp. 1109-1120, 2010

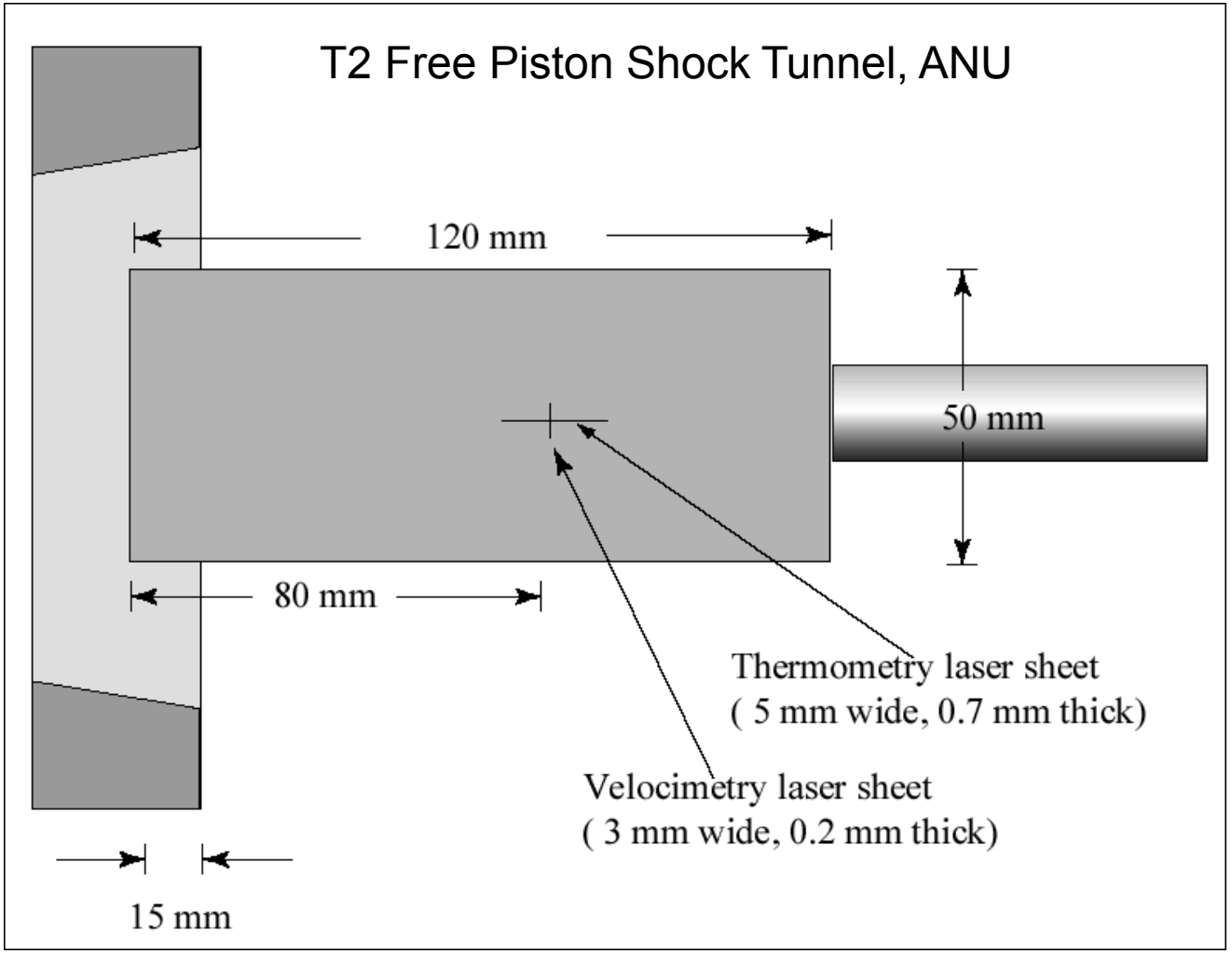
LIF Molecular Tagging Velocimetry (MTV)



- Boundary layer: slow moving gas near surface
  - Important: heat transfer, drag, separation, etc.
- Instantaneous measure of 1 velocity component
  - Can be extended to 2 or 3 components

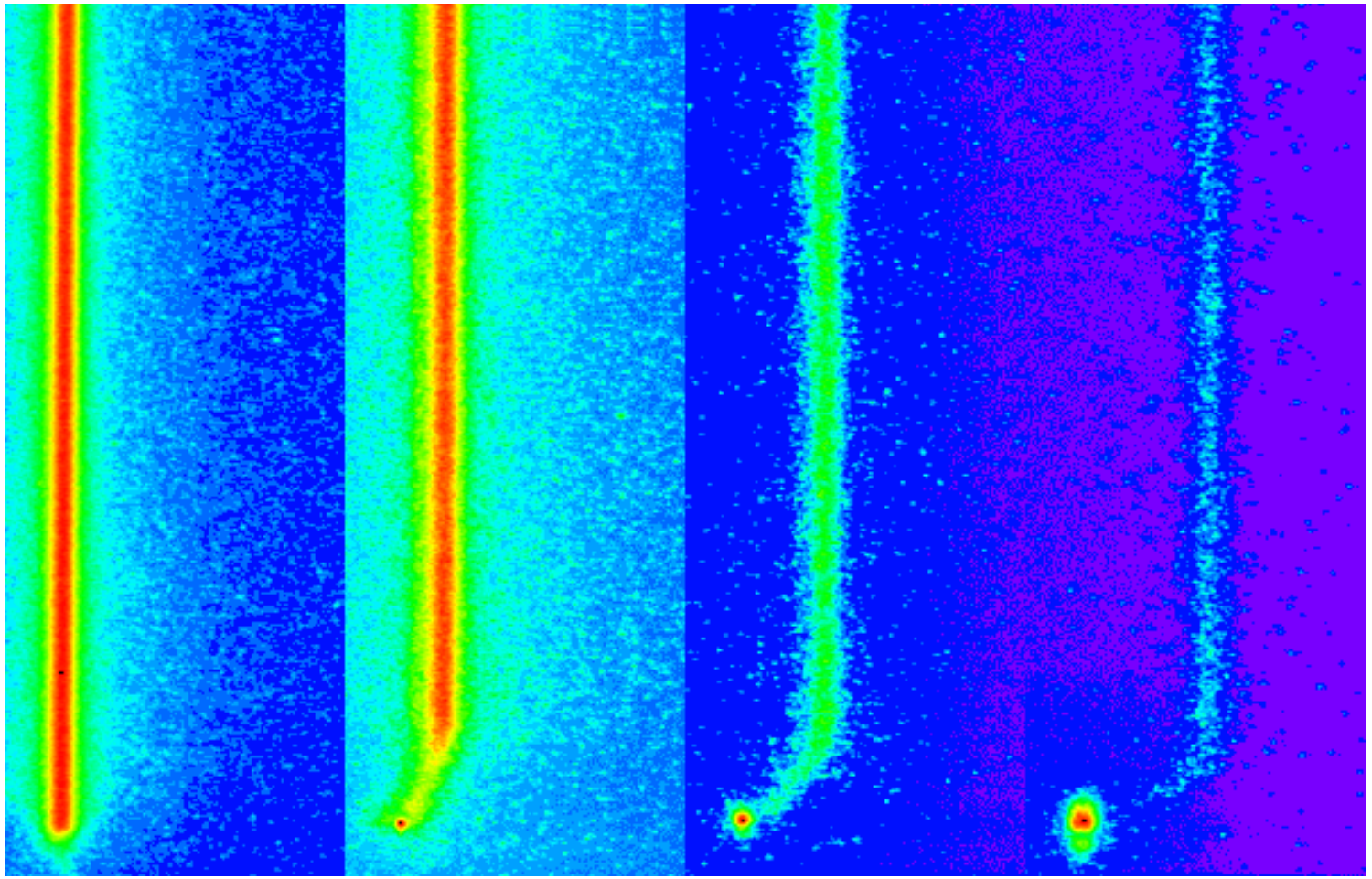
P. M. Danehy, S. O'Byrne, A. F. P. Houwing, J. S. Fox, and D. R. Smith, "*Flow-Tagging Velocimetry for Hypersonic Flows Using Fluorescence of Nitric Oxide*," AIAA Journal, 41(2), February 2003

Experimental Setup: Top View



Velocity ~ 3000 m/s; Mach Number = 7, laminar boundary layer

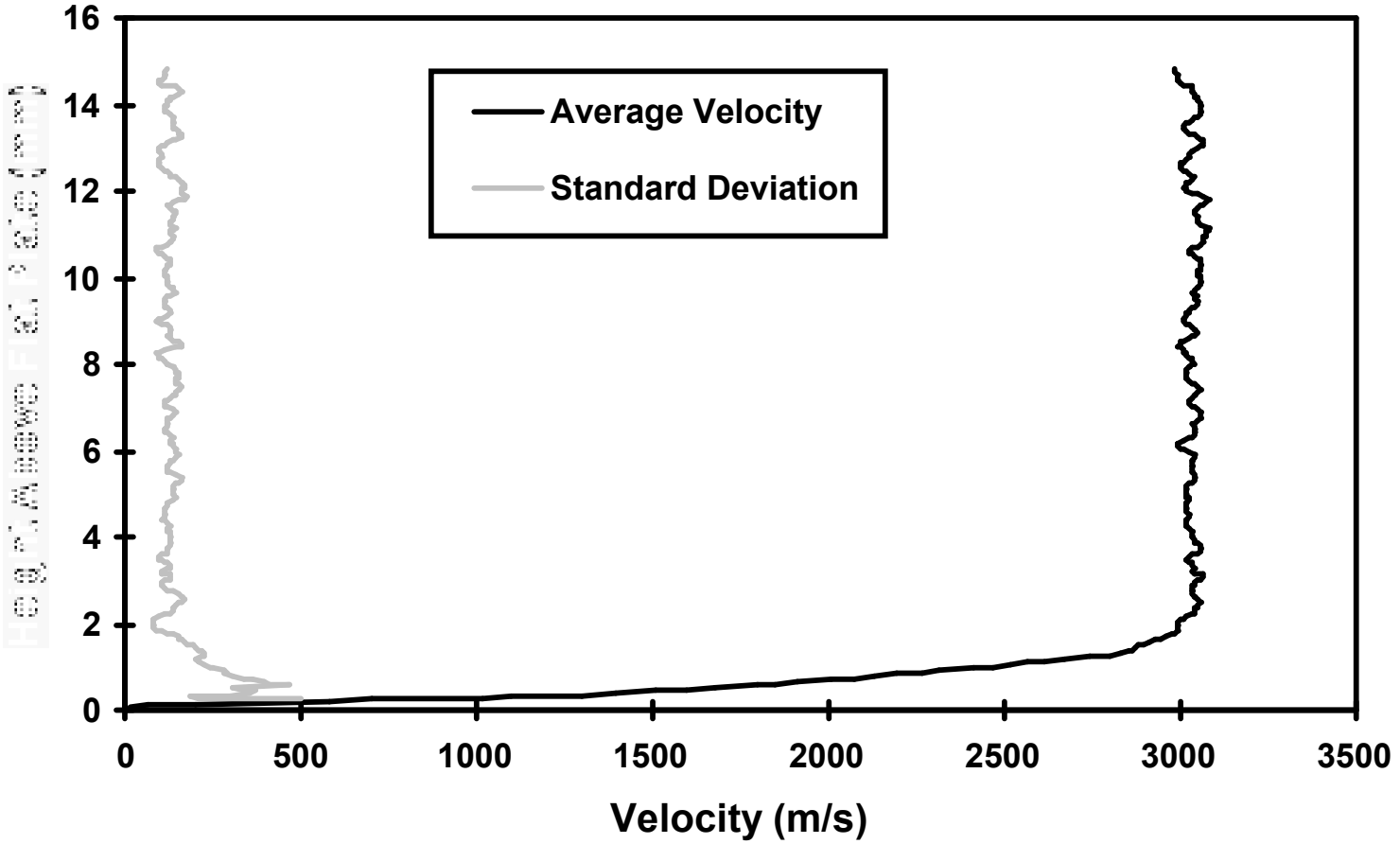
MTV: Delayed Images



Delay = 0 | 250 ns | 500 ns | 750 ns | ln(data) shown here

- Measurements performed on successive tunnel runs

Average of Single-Shot Measurements



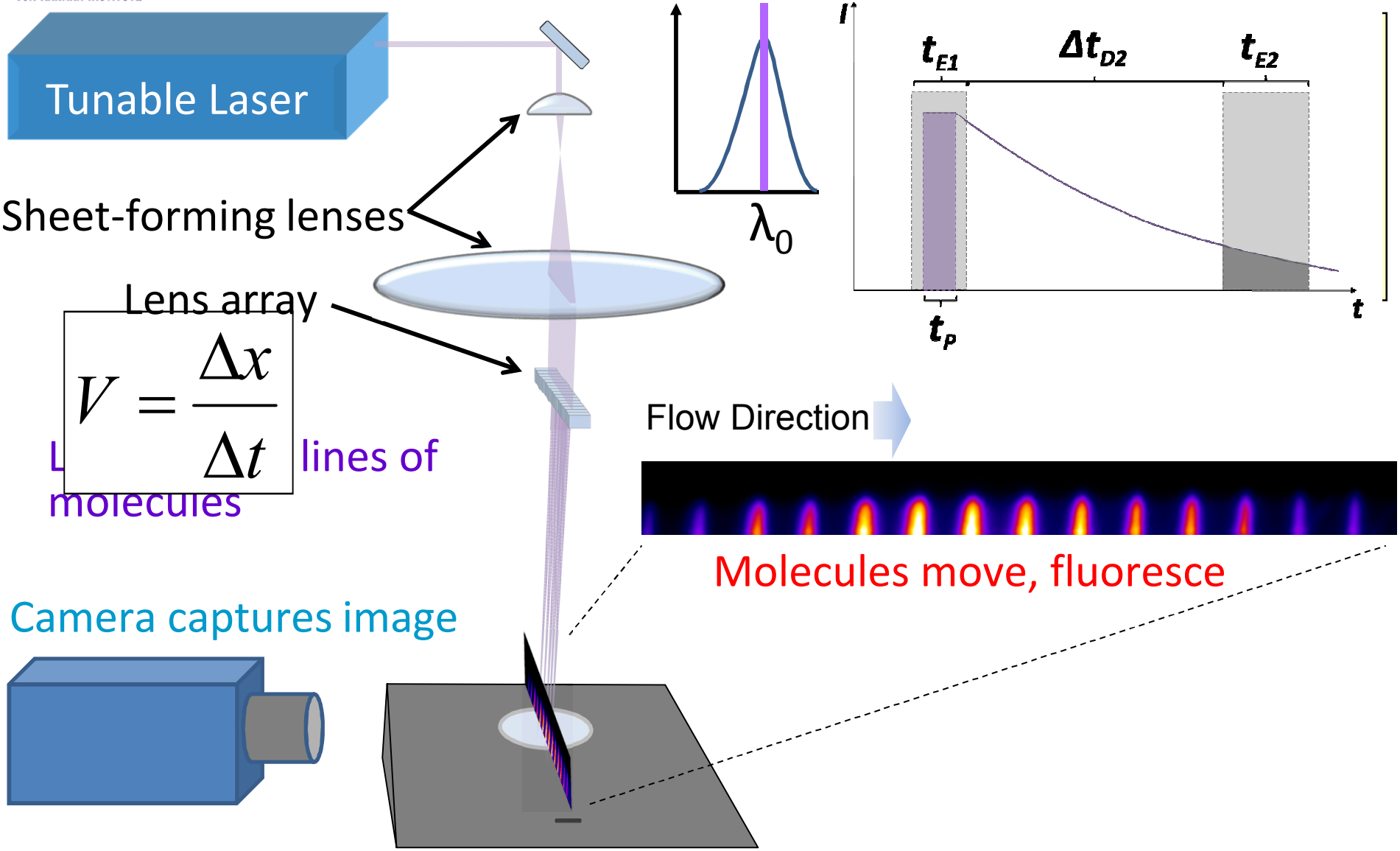
- Freestream velocity is uniform at 3035±60 m/s.
- 95% boundary layer thickness is 1.4±0.1 mm.
- Single shot measurement precision: ~60 m/s out of 3000 m/s (2%)
  - uncertainty: ~80 m/s (3%)

Single-Laser MTV

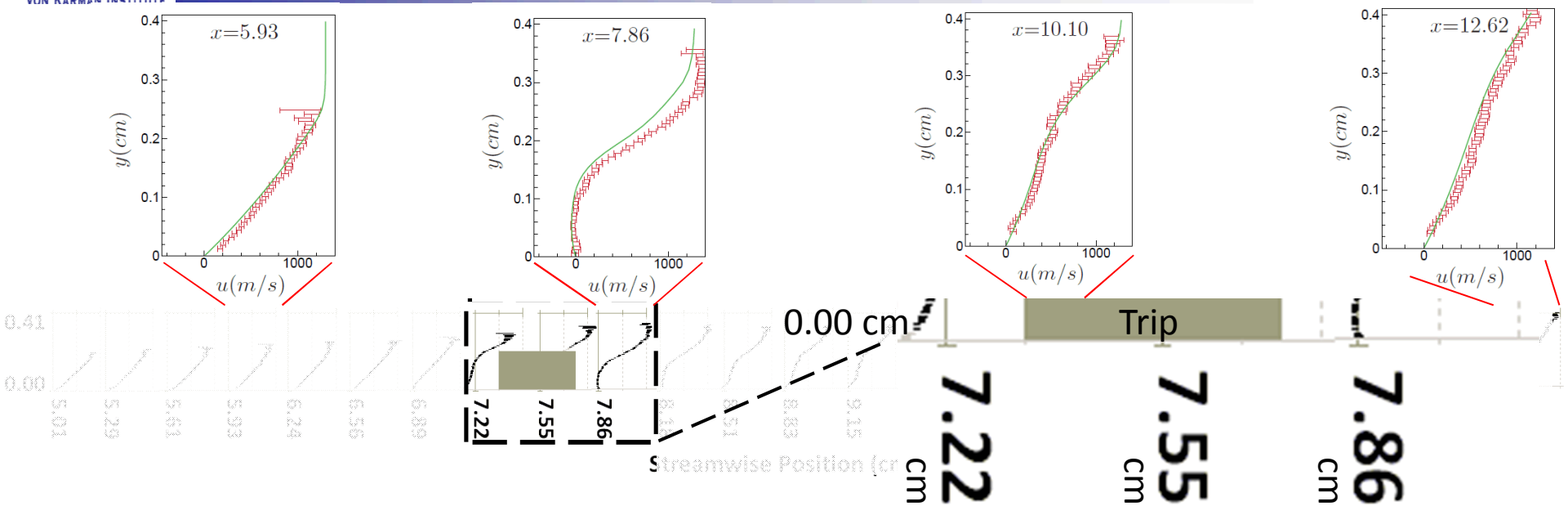
- Biacetyl (Hiller et al, 1984; Koochesfahani, 2007)
- Acetone (Lempert, 2002)
- NO (Danehy, 2003; Hsu, 2009; Bathel, 2011)
- I_2 (Balla, 2013)
- N_2 using FLEET (*Femtosecond laser electronic excitation tagging*) (Michael, 2011)
  - Does not require seeding
  - Does require an expensive fs laser

- *LIF lifetime must be long enough so molecules still emit light after (slightly) moved*

Single-laser MTV



Single-laser MTV



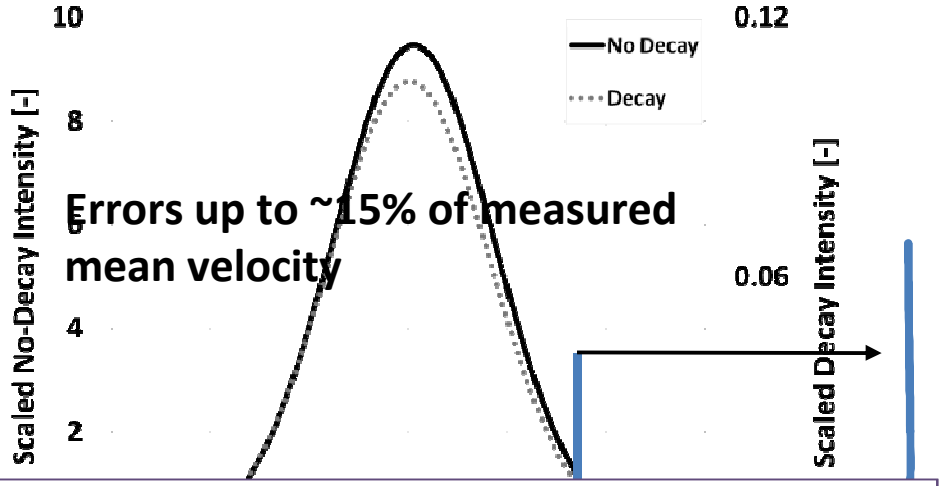
- **Velocity Measurement**
  - Single-component
  - Developed thorough uncertainty analysis methodology
  - Data used in subsequent CFD study by Iyer et al. of Univ. of Minnesota (AIAA 2011-566)

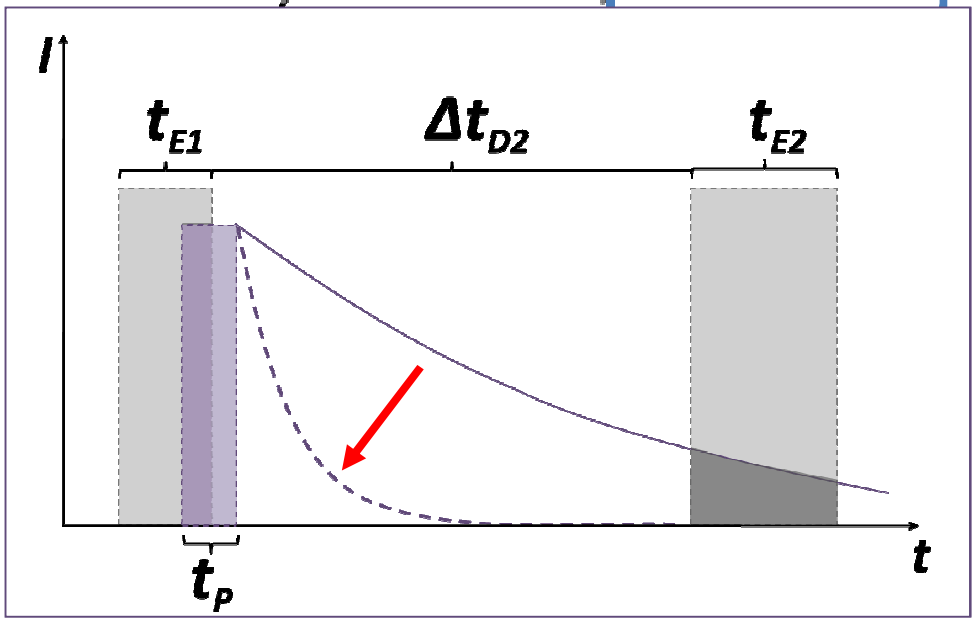
B. F. Bathel, P. M. Danehy, J. A. Inman, S. B. Jones, C. B. Ivey, C. P, and Goyne, "*Velocity Profile Measurements in Hypersonic Flows Using Sequentially Imaged Fluorescence-Based Molecular Tagging*," AIAA Journal, 49(9), September 2011

Single-laser MTV

- **Analysis**
  - Uncertainties in mean: as good as 3% of freestream (~1000 m/s) for 38 images
  - Modeled error from intensity decay, exposure duration, motion blur
  - Error from velocity component parallel to MTV profiles quantified

- **Study conclusions**
  - Velocity gradients can bias measurement
  - Scatter off of the model surface limited proximity of measurement to the wall
  - Limited to low-pressure conditions
  - Need higher pressure capability for higher Reynolds number required to study laminar-to-turbulent transition at least on this short model





Two (or Three) laser MTV

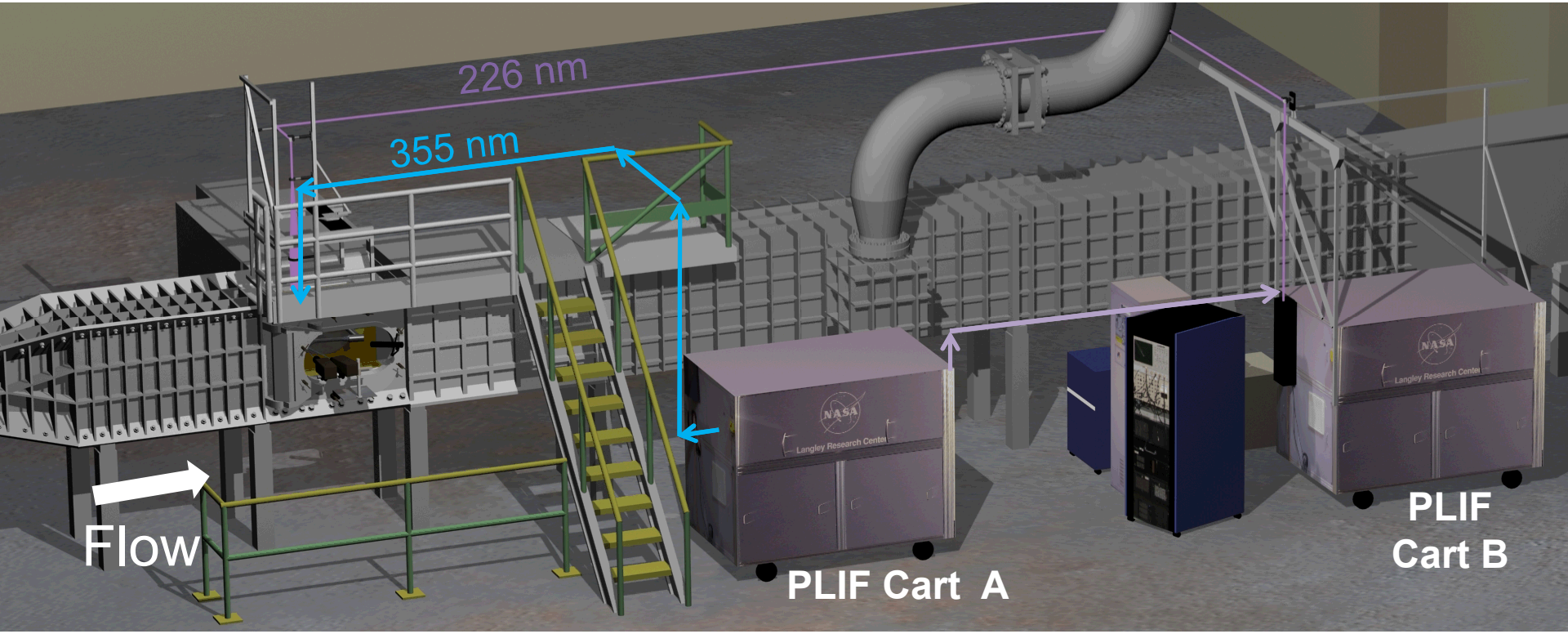
- Typically, one laser generates a line, series of lines or a grid pattern by, ionization (LEI[126-128]), vibrational excitation (RELIEF[129-131]), photolysis, bleaching or other process. Additional laser(s) read out pattern.
  - Example: Excimer laser at 193 nm+N_2+O_2→NO
  - *Air Photolysis and Recombination Tracking (APART)*
    - Dam et al, Optics Letters, 26 (1), pp. 36-38 2001
- A list of partner species used in the photolysis writing and reading process includes:

H_2O-OH,[133-139] N_2O-NO,[140] O_2-O_3,[141,142]
N_2/O_2-NO,[143-146] and NO_2-NO.[116,145-154]

- Method can use air constituents (N_2/O_2, O_2, N_2) or can be seeded (H_2O, N_2O, NO_2, Na, etc.).
  - We have used NO_2→NO to study transition

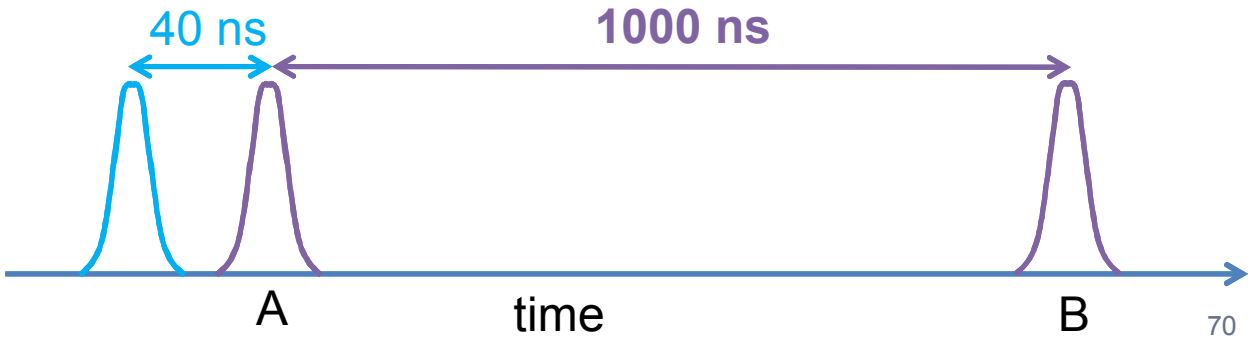
Three-Laser MTV (NO_2→NO velocimetry)

(for details of velocimetry method, see Bathel et al, AIAA-2011-3246 and others incl. TAMU[149], OSU[150], etc.)



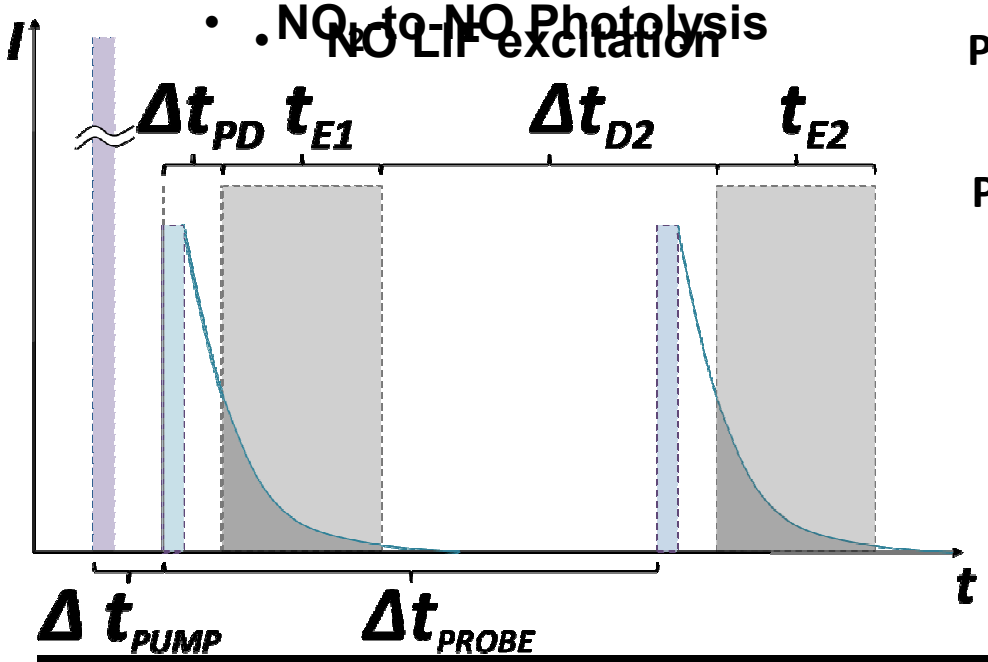
NO_2+355 nm → NO

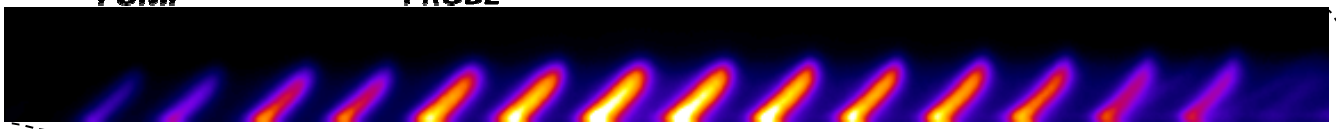
Write lines of NO into NO_2 seeded in the flow



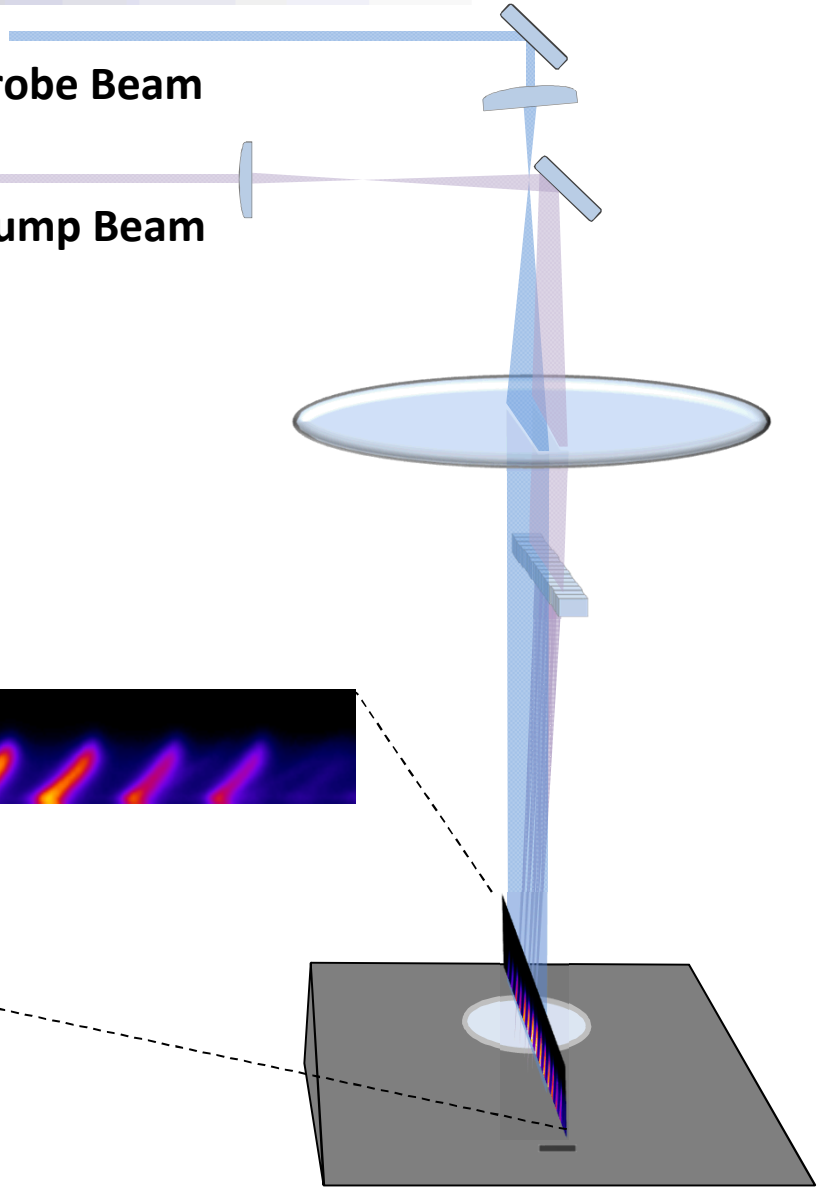
Three-laser MTV

- NO_2 to NO Photolysis
- NO LIF excitation

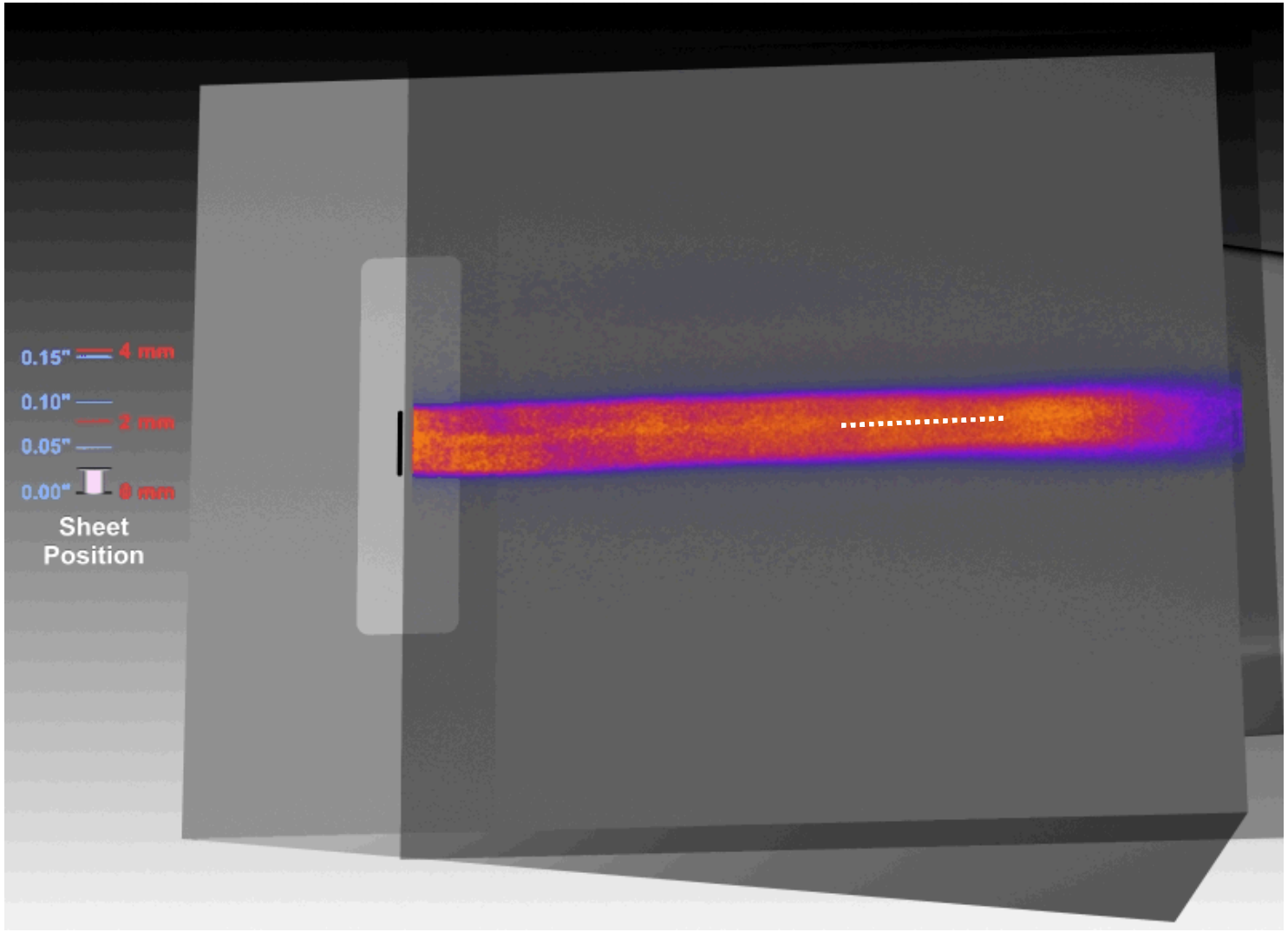








Flow Visualization: No trip

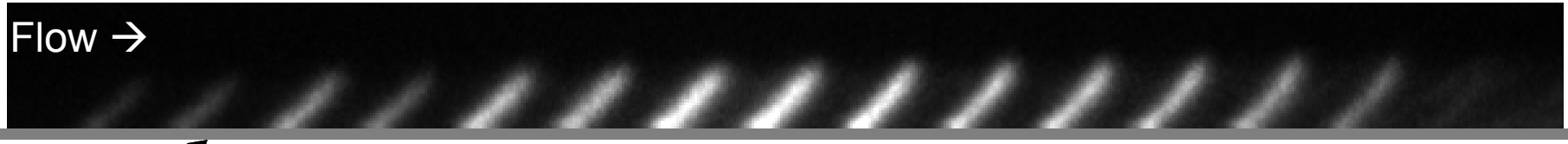


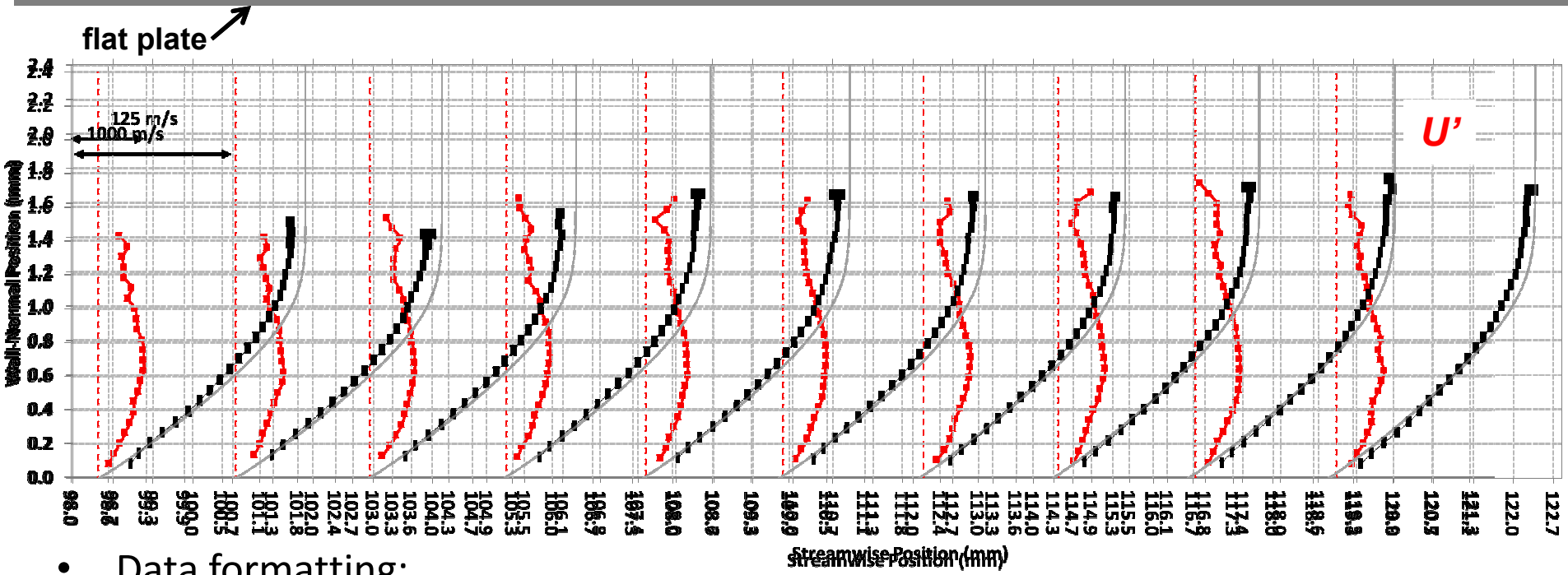
PA = 20°
mid-Re

$M_e \approx 4.2$

- Flow appears laminar when no trip is present

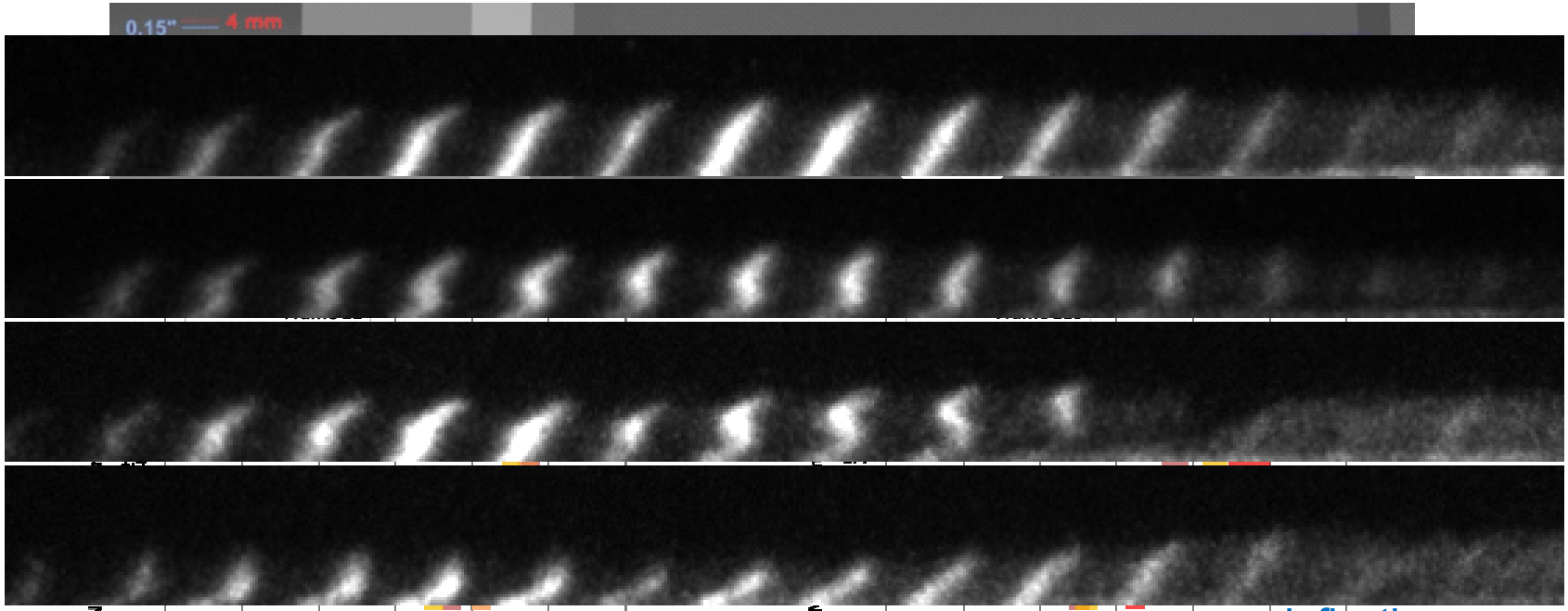
Velocimetry: No Trip

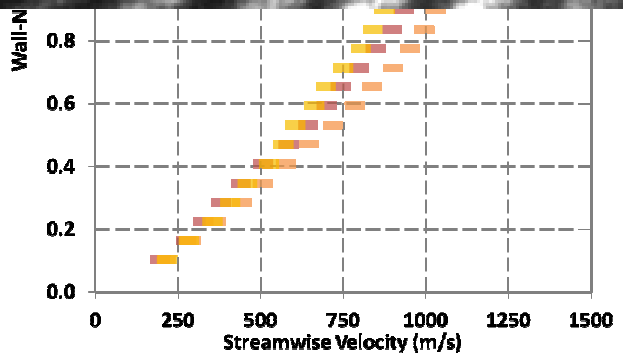


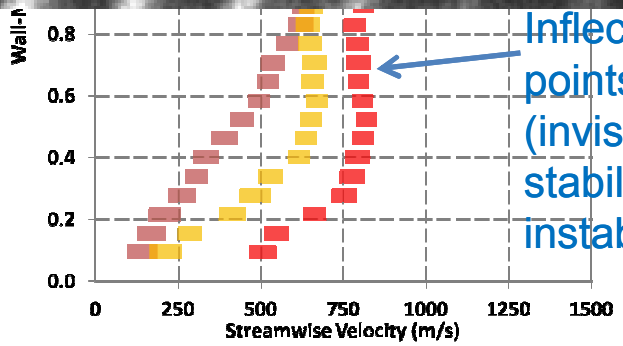


- Data formatting:
  - Center of the data point is mean velocity; Width is uncertainty in mean
  - Standard deviations shown in red (instrument error ~2% of max U was subtracted)
- Blasius-like boundary layer profiles observed (to within 100 μm of wall)
  - Deficit near edge is likely caused by seeding in zero velocity gas into BL (see Johansen et al)
  - U' ~ 80 m/s at maximum. Peak U' is ~ 0.7 mm above surface.

Velocimetry: 1-mm Tall Trip (centerline)



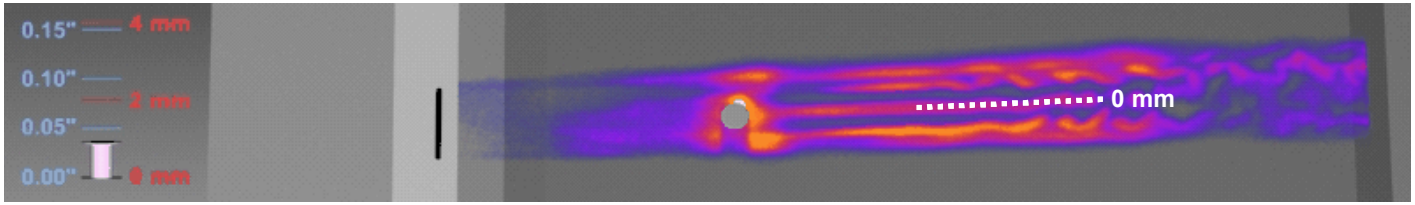


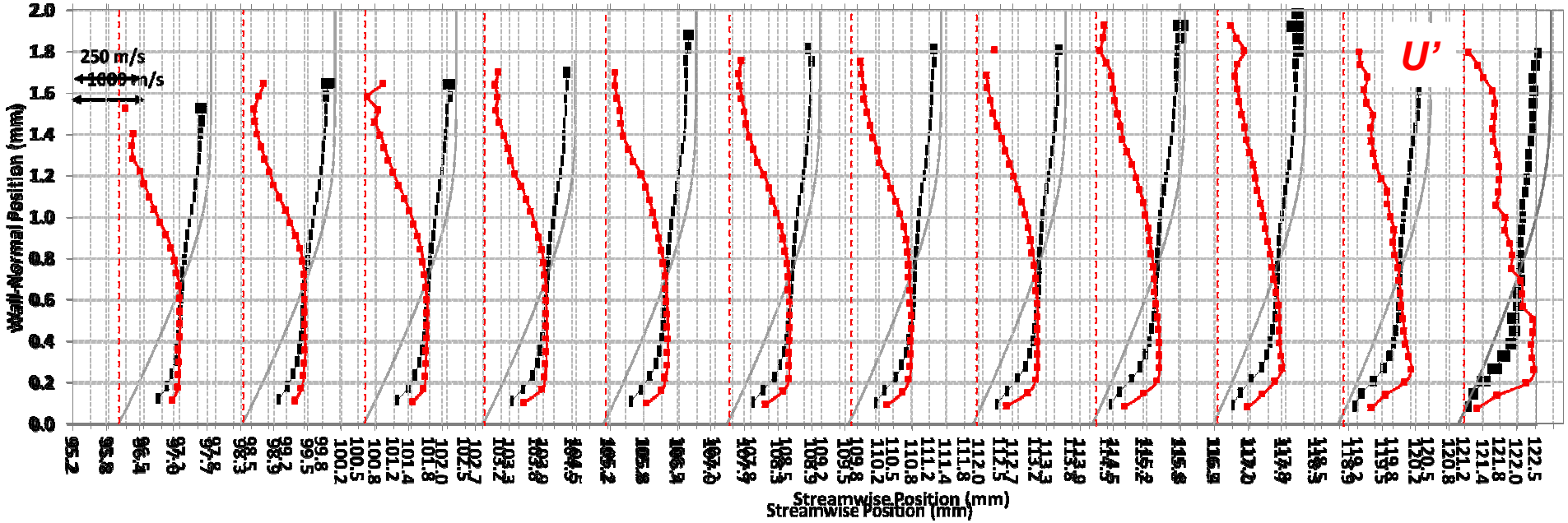


Inflection points (inviscid stability/ instability)

- See many "S" shaped profiles in single shots, also in mean
  - Strong profile-to-profile correlation *within* one image.
  - Seen in prior laminar measurements (Bathel et al, Paper AIAA-2010-4998 different Mach)
- Tripped flow fluctuates much more than untripped flow

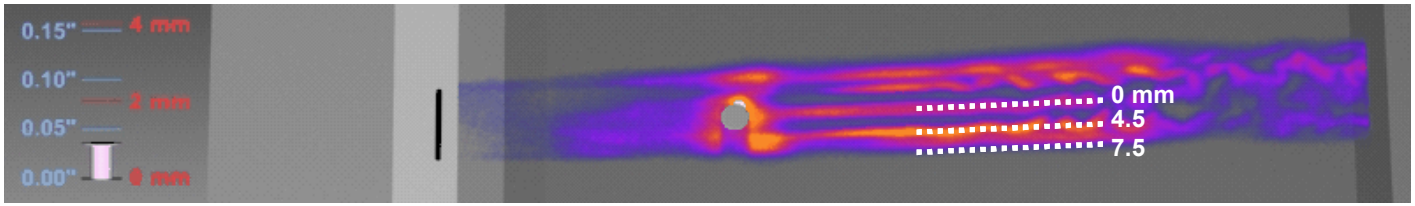
Velocimetry: 1-mm Tall Trip (centerline)

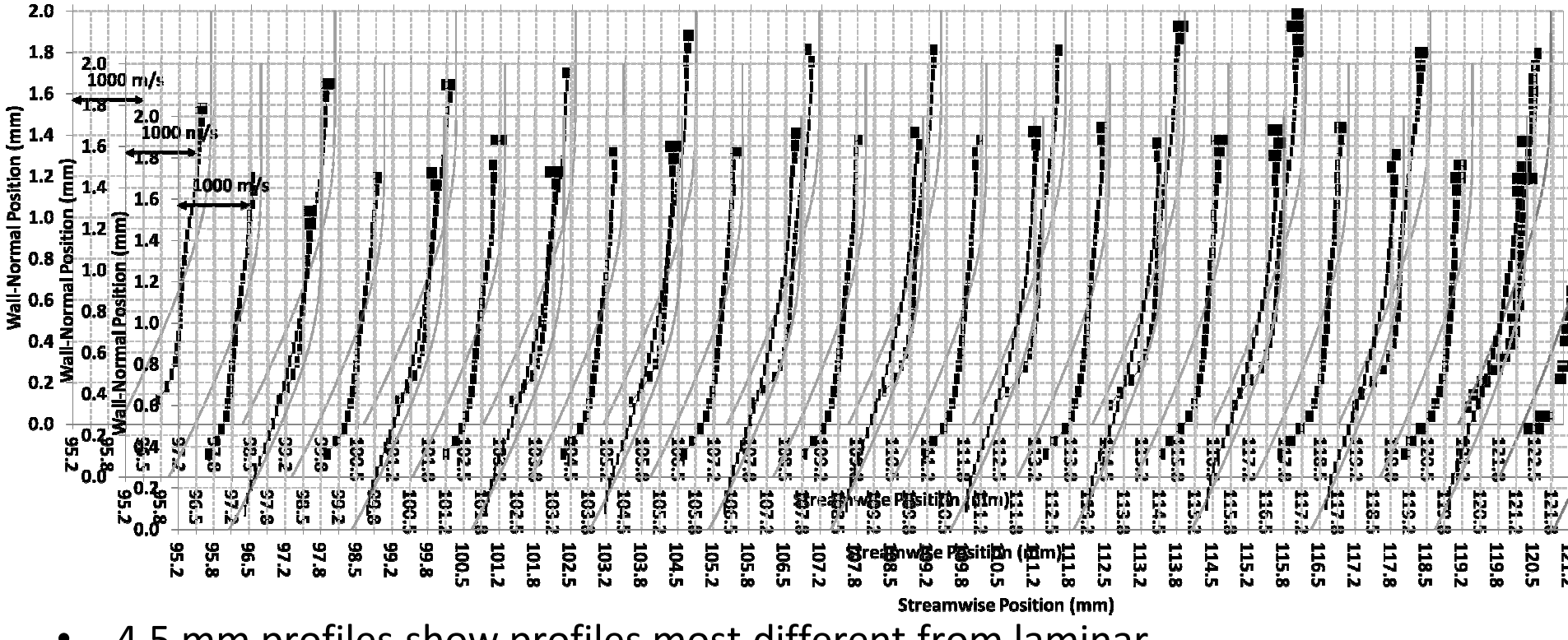




- Velocity profile is full near flat plate with a deficit near edge of BL
  - Note: 2x lower flowrate in this run compared to no-trip case previously shown.
- U' data has increased to peak ~250 m/s (3x larger than laminar case).
- Shape of U' profile changes slightly with distance downstream.

Velocimetry: 1-mm Tall Trip





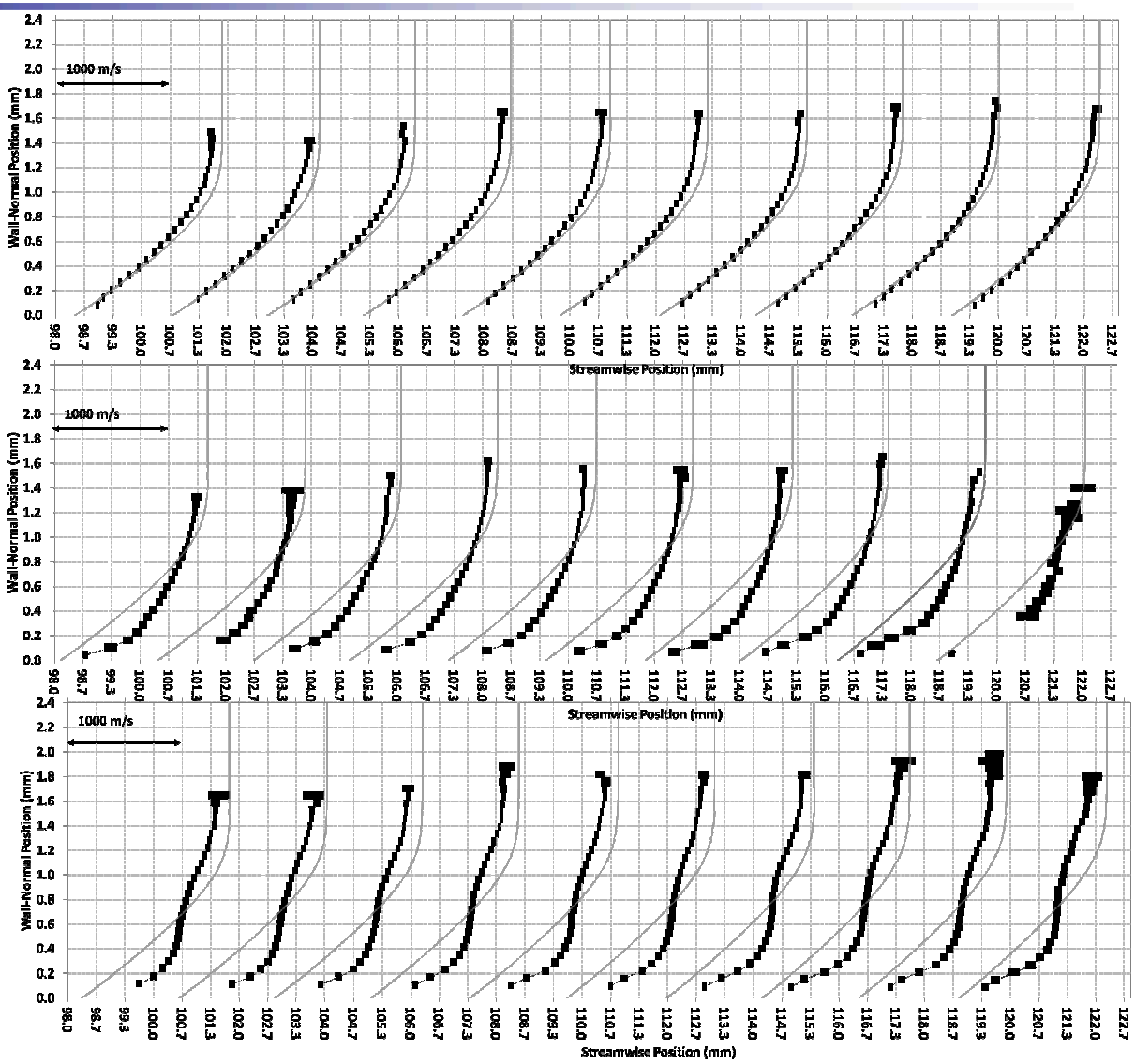
- 4.5 mm profiles show profiles most different from laminar
- Flow becomes more laminar far from centerline (similar to flow vis)
  - U' drops back to ~80 m/s

Comparison of *U* for different trip heights

No Trip

0.53-mm Trip

1.0-mm Trip



measurements on centerline

$Re_{kk} \approx 400$

$Re_{kk} \approx 2000$

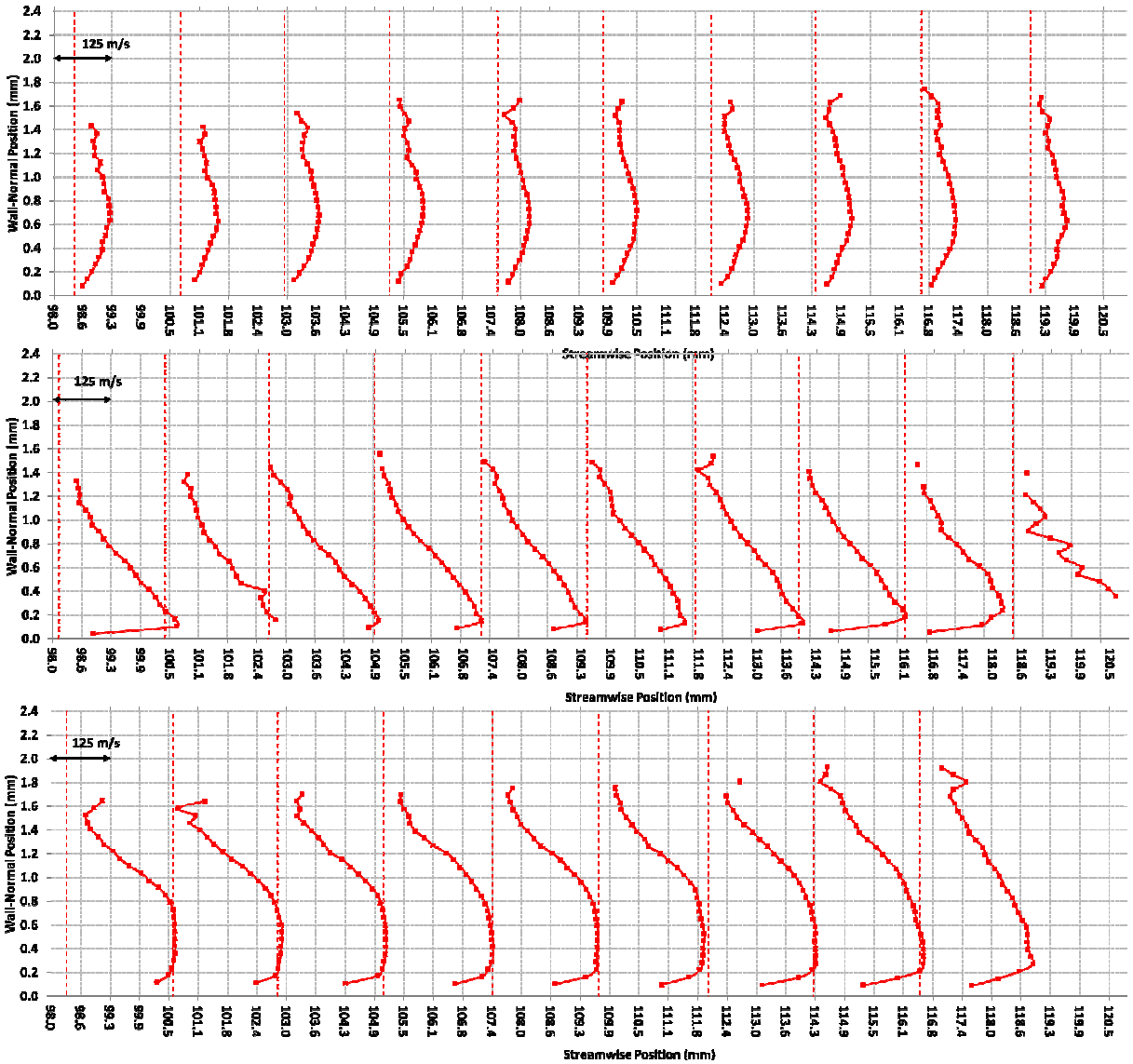
- 0.53 mm trip profile is between no and 1-mm but no inflection point.

Comparison of *U'* for different trip heights

No Trip

0.53-mm Trip

1.0-mm Trip



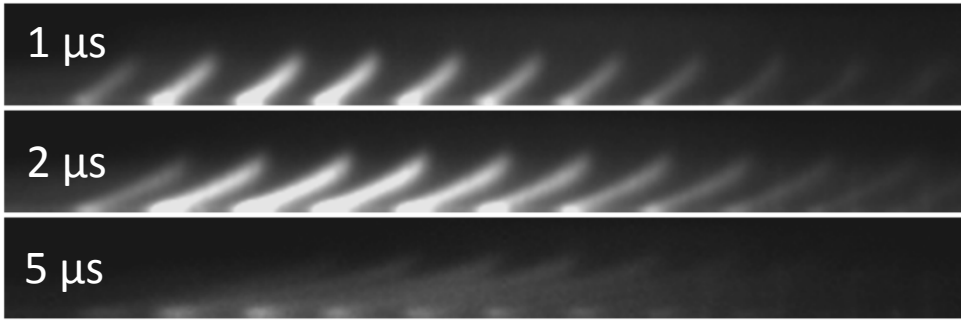
measurements on centerline

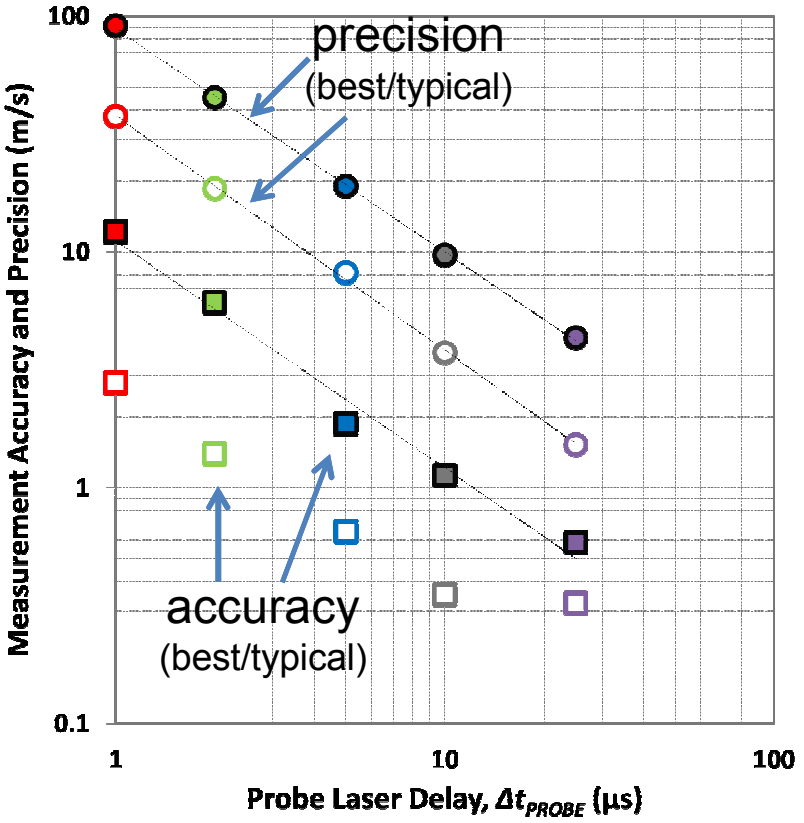
- 0.53 mm trip *U'* peak much closer to plate.

Three-laser MTV: uncertainty & design

- **Analysis**
  - Uncertainty for high s/n data
    - Mean: 1% of edge velocity (~1300 m/s) in 200 images
    - Single-shot: 1-2% of edge velocity
  - Measurement within 100 μm from wall

- **Tradeoffs for experiment design**
  - Accuracy and precision improve with probe delay
  - Spatial resolution worse with longer probe delay
    - Can use longer delay for slow flow
  - Current configuration requires relatively large blowing rates for adequate signal-to-noise
    - May be significant wall-normal velocity component

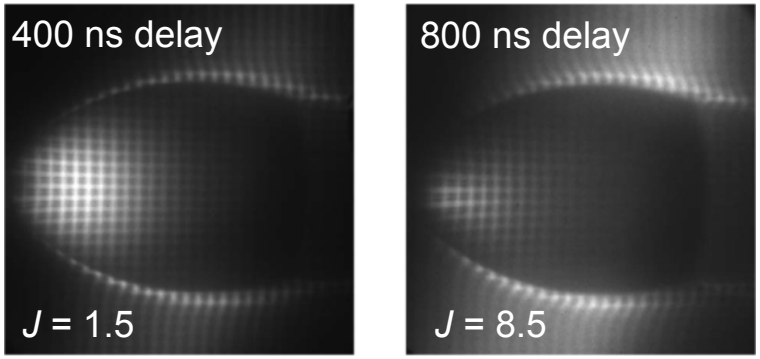




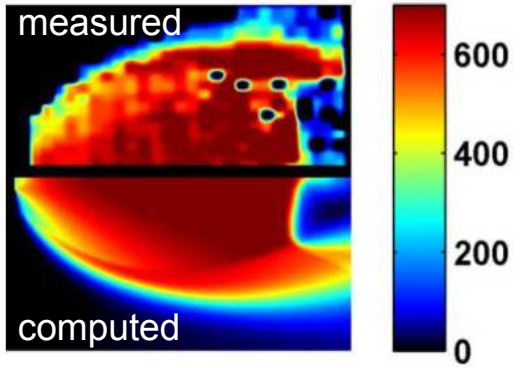
Data obtained at static, low pressure conditions

3 laser MTV: measure u, v, T

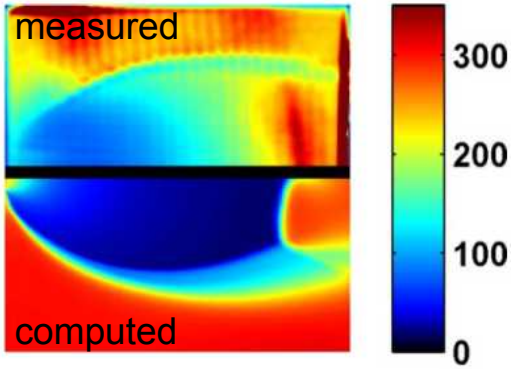
- Texas A&M has used 3 laser technique with crossed beam pattern for 2-component velocity and temperature in an underexpanded N_2 jet flow with NO_2:
  - VENOM: vibrationally excited nitric oxide monitoring
  - Excite to v' = 1 state



Raw data images



U-Velocity (m/s)
(~5% uncertainty)

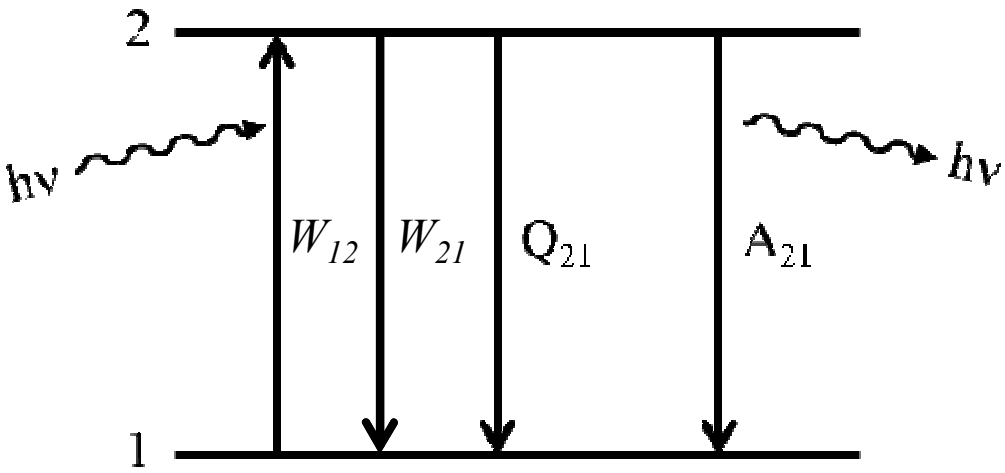


Temperature (m/s)
(~9-35% uncertainty)

R. Sánchez-González, R. Srinivasan, R. D. W. Bowersox, and S. W. North, "*Simultaneous velocity and temperature measurements in gaseous flowfields using the vibrationally excited nitric oxide monitoring technique: a comprehensive study*," Applied Optics, 51(9), pp. 1216-1228, March 20, 2012

Species Concentrations

- Problem: How to deal with unknown Q_{21}?
- Solutions:
  - Saturated LIF: $(W_{12} + W_{21}) \gg (Q_{21} + A_{21})$
    - Q_{21} is negligible compared to W's
    - Hard to achieve
  - Predissociation
    - $Q_{pre} >> Q_{21}$
  - Measure τ_{LIF}
    - Determine Q_{21} from τ_{LIF}
  - Measure other species present and predict τ_{LIF}
    - Raman/Rayleigh/LIF (Sandia and DLR)
  - Counter-propagating beams, take ratio (Versluis, 1997)
- Potential problems: absorption, rad. trapping

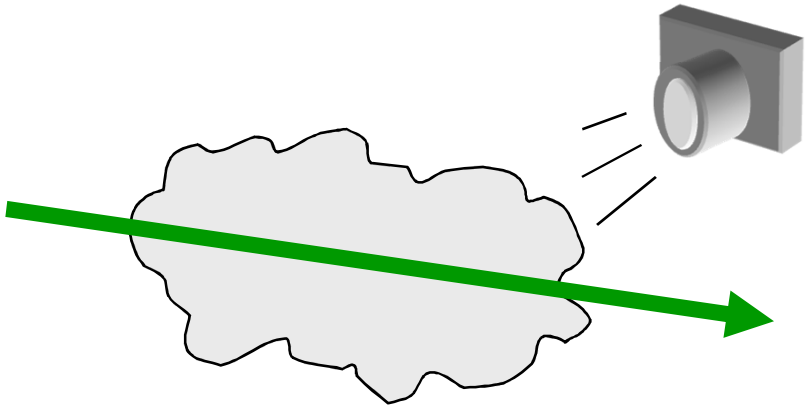


LIF Advantages/Disadvantages

- Advantages
  - Relatively easy to make planar measurements (PLIF)
  - Provides flow visualization simultaneously
  - Relatively easy to perform
  - Can probe many different parameters (T, V, concentration)
    - Will show velocity measurements with 1-2% precision
- Disadvantages
  - Precision of single-shot temperature measurements (~20%) is not good compared to CARS (2-3%) or LITA (1%)
  - Difficult to do quantitative concentration measurements, especially planar

Spontaneous Rayleigh and Raman Scattering

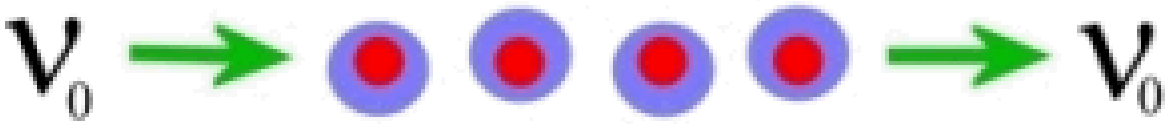
Rayleigh and Raman Scattering



- When light passes through a gas:
  - LIF is a *resonant* absorption/emission process
  - Mie, Rayleigh and Raman are *scattering* processes
    - Mie Scattering: $d \geq \lambda$ (d is the particle size)
    - Spontaneous Rayleigh and Raman scattering: $d << \lambda$
- Will explain Rayleigh and Raman two ways
  - Physical interpretation
  - Classical description
  - Not going to discuss quantum treatment

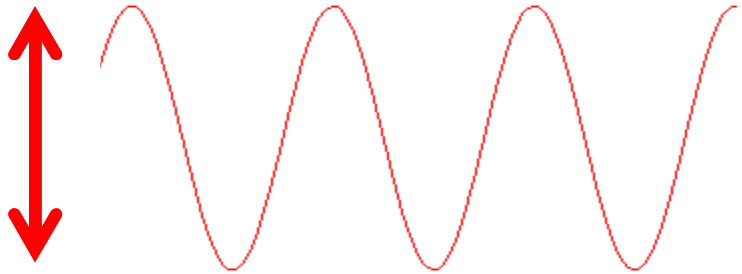
Rayleigh Scattering

- Rayleigh scattering:
  - Electric field causes electron cloud to oscillate with respect to the nucleus: induced dipole



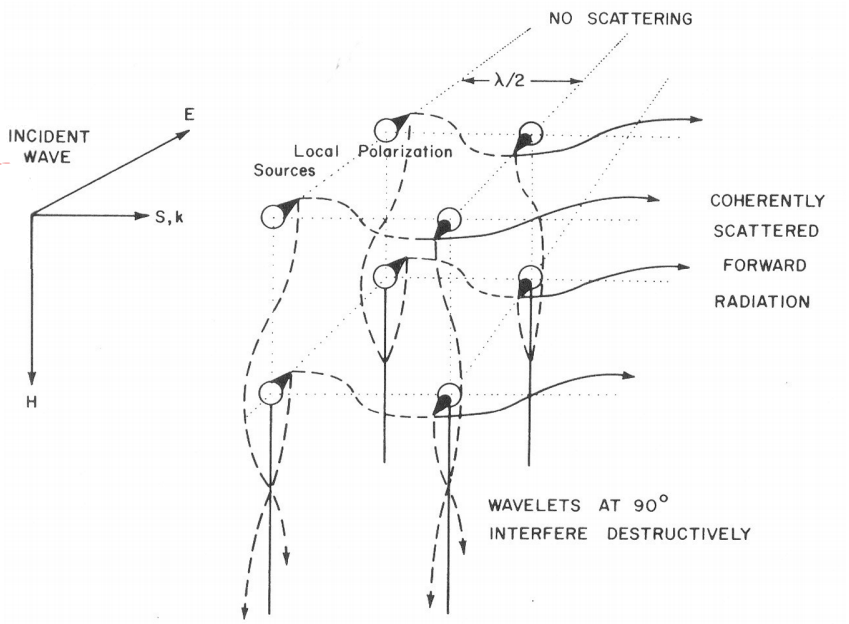
http://www.timkelf.com/Research/ResearchSERS.html

  - An oscillating dipole acts like an antenna: radiates!
    - Radiation is perpendicular to oscillation



Many Particle Scattering

- A plane-wave perturbs many particles: they radiate
  - Interfere constructively in direction of propagation
  - Interfere destructively in other directions

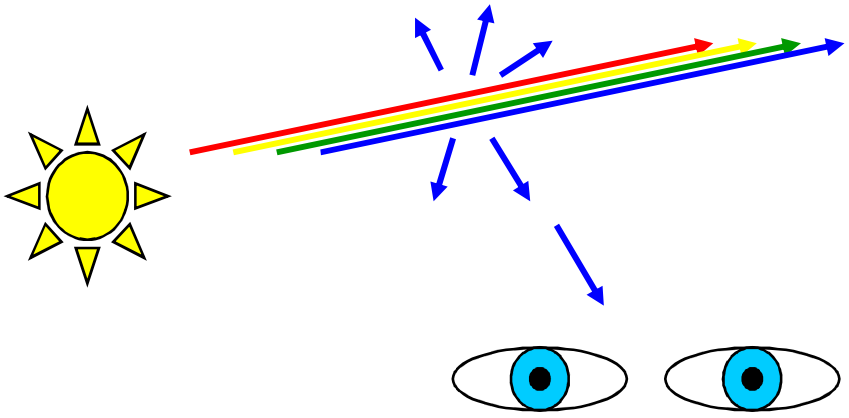


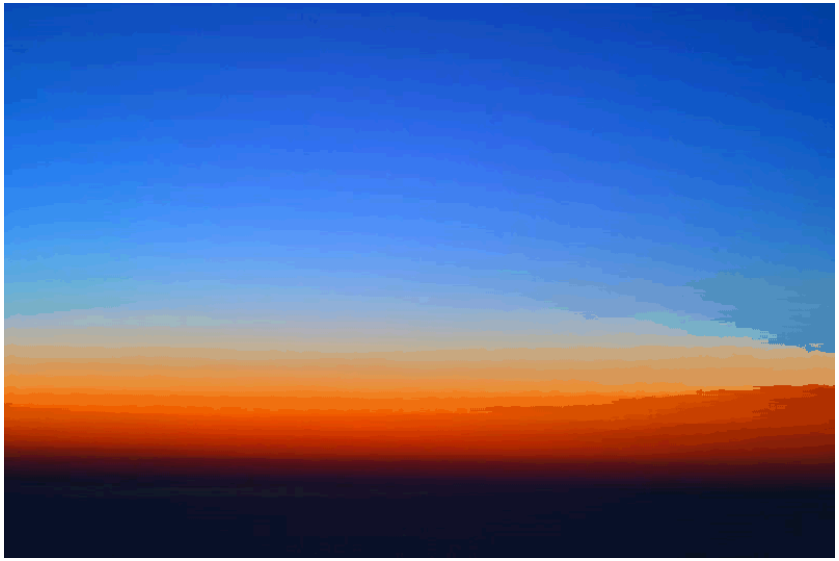
G.C. Baldwin "An introduction to nonlinear optics." *New York: Plenum Press*, (1969).

  - If same number of particles everywhere in gas, destructive interference would give no side scatter
    - But random statistical variation in particle concentration: deconstructive interference is incomplete: Rayleigh Scattering!

Rayleigh Scattering

- Very weak process
  - 1 photon in 0.1 million scatters / meter at STP
- Wavelength dependence: $1/\lambda^4$



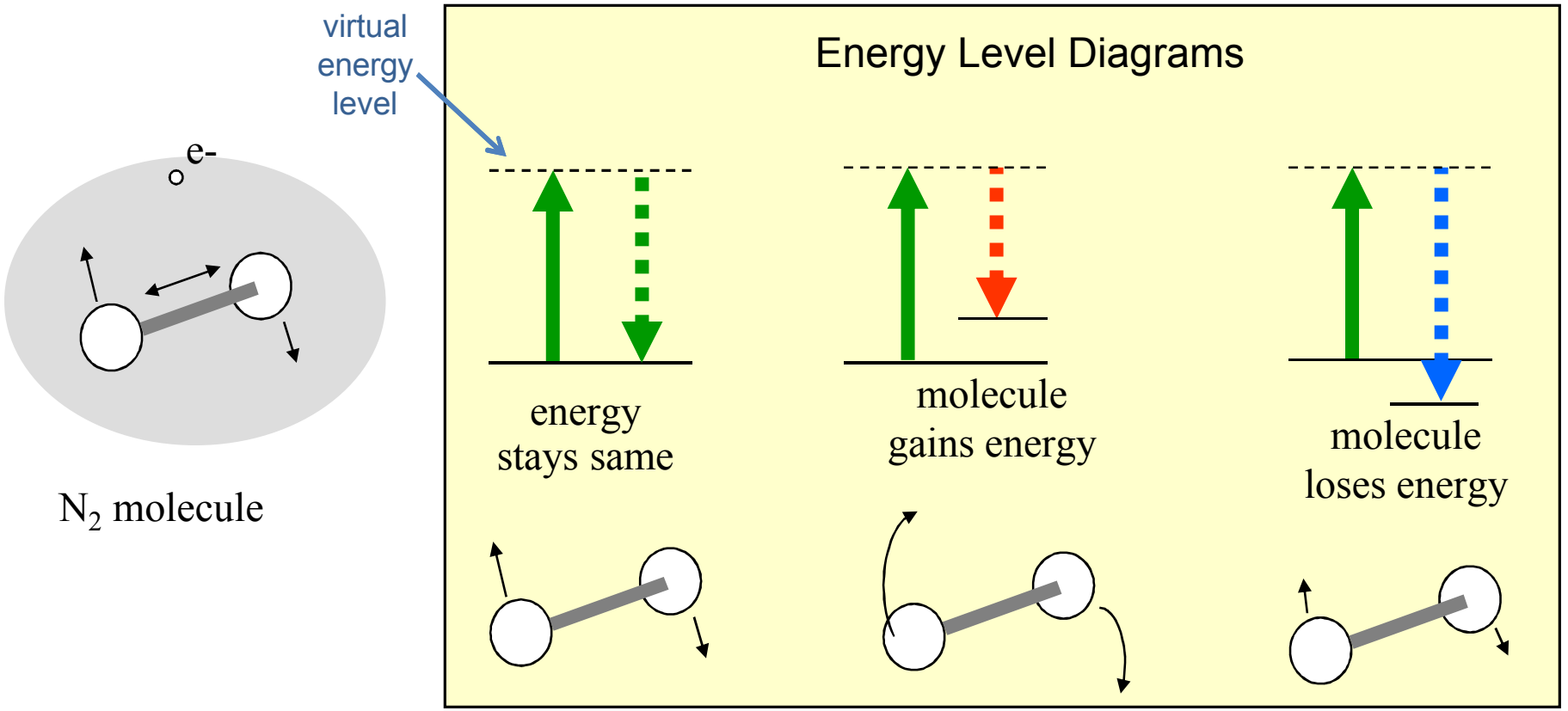


Rayleigh scattering in the atmosphere after sunset, picture taken over the ocean, at 500m altitude.
http://en.wikipedia.org/wiki/Rayleigh_scattering

- Causes blue sky and red sunsets
  - Mie scattering from aerosols also contributes

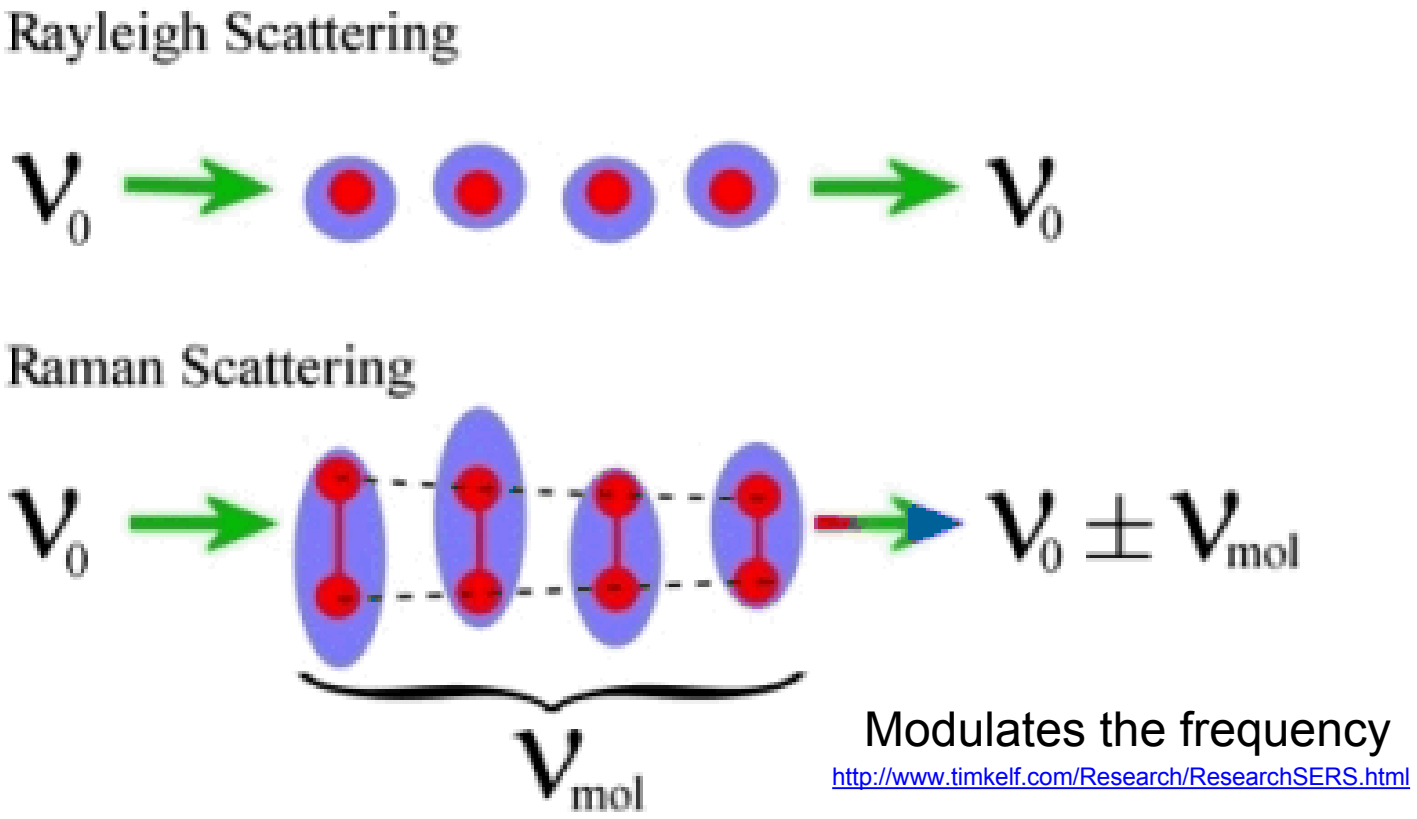
Raman Scattering

- Similar to Rayleigh scattering but molecules gain or lose energy during scattering process
- Raman x1000 lower probability than Rayleigh



Raman Scattering

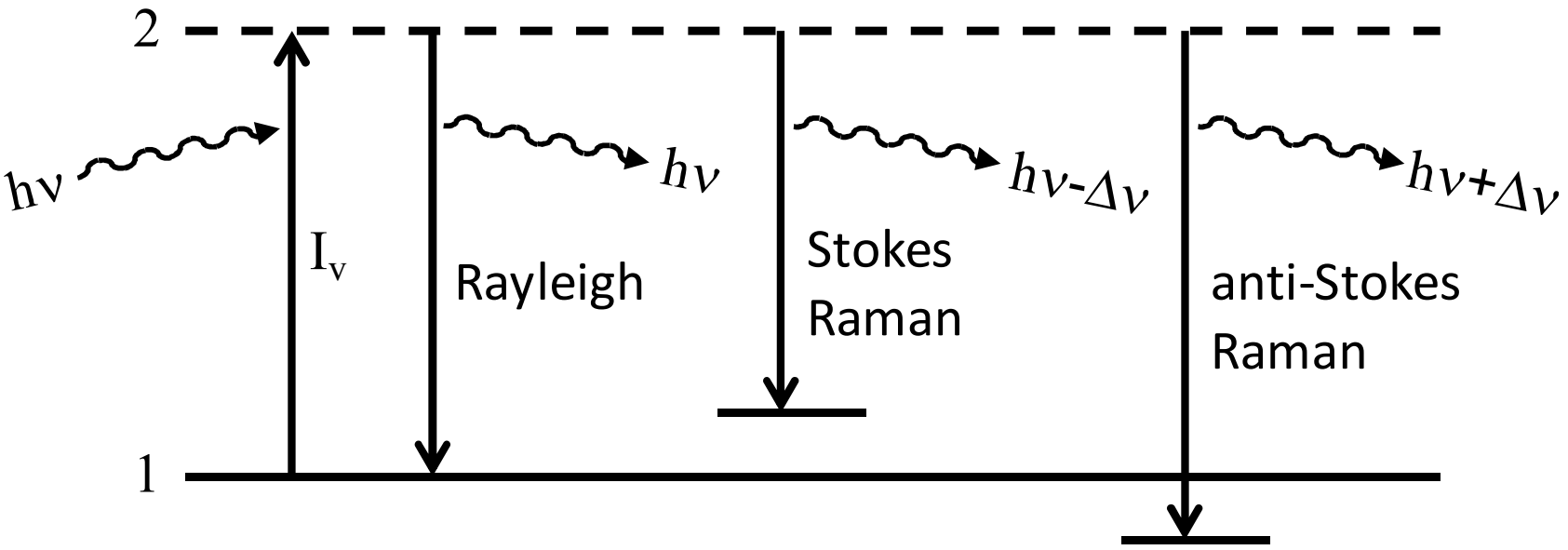
- Raman scattering results in a frequency shift
  - Molecule vibrating at frequency ν_{mol}



Modulates the frequency

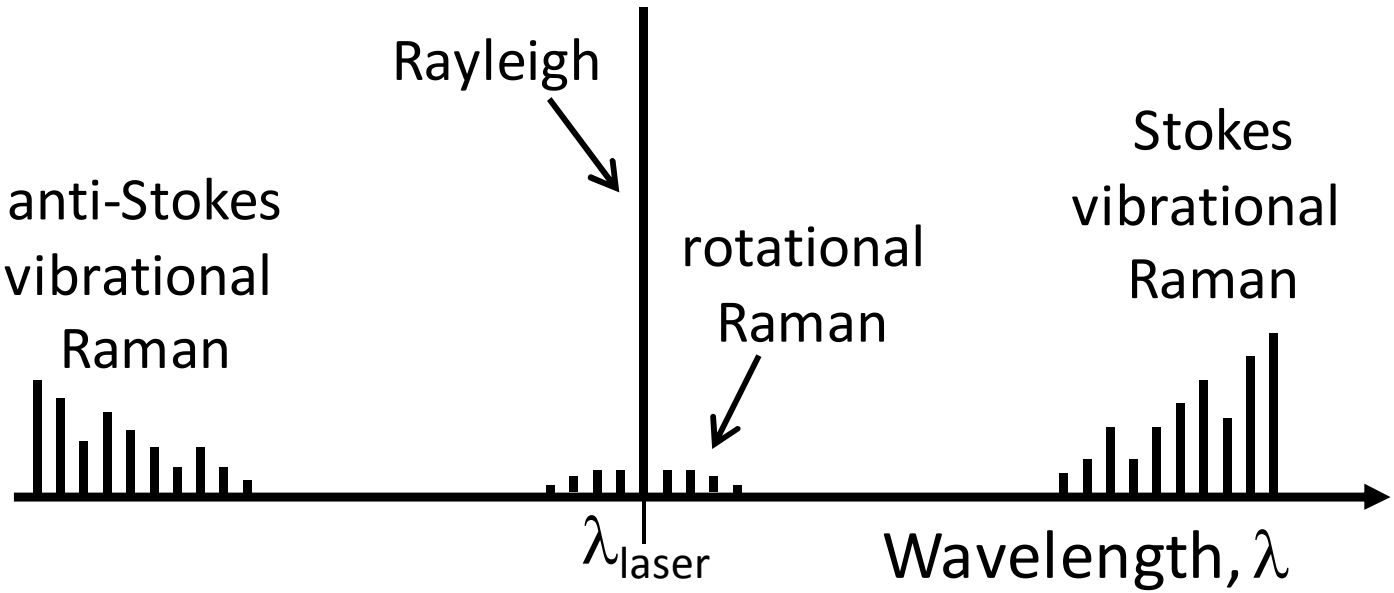
http://www.timkelf.com/Research/ResearchSERS.html

Rayleigh and Raman Scattering



- Energy level diagram:
  - Stokes Raman: energy absorbed by molecule
  - anti-Stokes Raman: energy lost by molecule
    - Initial state must not have been a ground state

Rayleigh and Raman Spectra



- Rayleigh: near laser's wavelength
- Raman: frequency shift depends on molecule, state (sensitive to concentration and temperature)
  - Rotational Raman
  - Vibrational Raman

Rayleigh and Raman Theory 1

- Scattered intensity is proportional to the square of the induced dipole moment, $\vec{p}$:

$$\vec{p} = \varepsilon_0 \alpha \vec{E}$$

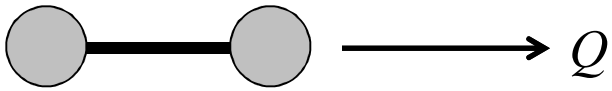
where ε_0 is the permittivity of free space, α is the molecular polarizability and $\vec{E}$ is the incident electric field given by:

$$\vec{E} = \overrightarrow{E_0} \cos(\omega_0 t)$$

where $\overrightarrow{E_0}$ is the amplitude of the electric field, ω_0 is the frequency of the laser light and t is time.

Rayleigh and Raman Theory 2

- Polarizability, α, depends on internal structure
  - varies with time during vibrational oscillations at the natural frequency of the molecule, ω_v



  - α varies with spatial coordinate, Q
    - More loosely held electrons → more polarizable
    - Taylor Series Expansion:

$$\alpha = \alpha_0 + \left(\frac{\partial \alpha}{\partial Q}\right)_0 dQ$$

where dQ is small displacement about equilibrium positions which occurs during normal vibration

$$dQ = Q_0 \cos(\omega_v t)$$

Rayleigh and Raman Theory 3

- Substituting previous equations into $\vec{p} = \varepsilon_0 \alpha \vec{E}$:

$$\vec{p} = \left[\alpha_0 + \left(\frac{\partial \alpha}{\partial Q}\right)_0 Q_0 \cos(\omega_v t)\right] \varepsilon_0 \overrightarrow{E_0} \cos(\omega_0 t)$$

- Expanding and using trig identity:

$$\vec{p} = \alpha_0 \varepsilon_0 \overrightarrow{E_0} \cos(\omega_0 t) + \tfrac{1}{2}\left(\frac{\partial \alpha}{\partial Q}\right)_0 \varepsilon_0 Q_0 \overrightarrow{E_0} [\cos(\omega_0 - \omega_v)t + \cos(\omega_0 + \omega_v)t]$$

Rayleigh Scattering — Stokes Raman — anti-Stokes Raman

- Resulting induced dipole moment (and scattering) is at three frequencies
- Amplitude is usually determined empirically:

$$\left(\frac{\partial \sigma}{\partial \Omega}\right)_{zz} \equiv \frac{I_{zz}^{\Omega}}{NI}$$

Differential Cross Section

(varies by molecule and with I but not with P, T)

(zz indicates polarization of laser and detection)

$$I_{zz}^{\Omega} = \left(\frac{\partial \sigma}{\partial \Omega}\right)_{zz} NI$$

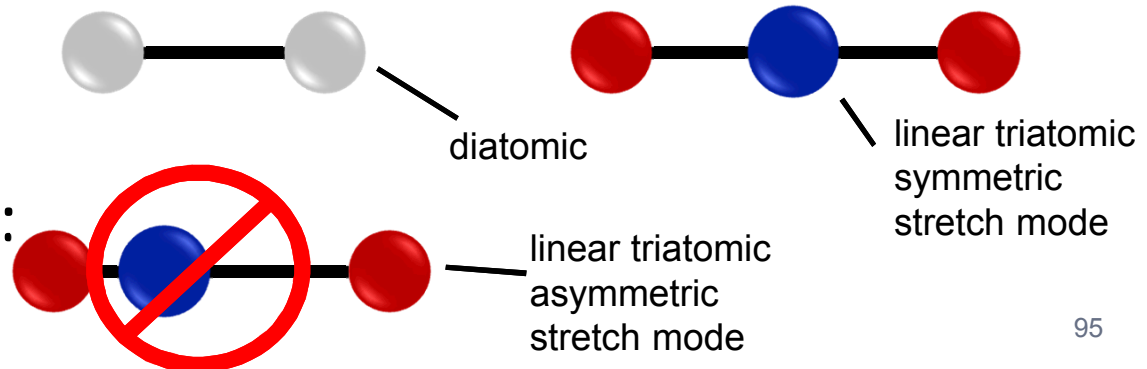
- I_{zz}^{Ω}: scattered power per solid angle
- $\left(\frac{\partial \sigma}{\partial \Omega}\right)_{zz}$: diff. cross section
- N: number density
- I: laser irradiance

Rayleigh and Raman Theory 4

- Selection Rules: Raman Activity
  - Not every molecule or vibrational mode produces Raman

$$\vec{p} = \alpha_0 \varepsilon_0 \overrightarrow{E_0} \cos(\omega_0 t) + \frac{1}{2}\left(\frac{\partial \alpha}{\partial Q}\right)_0 \varepsilon_0 Q_0 \overrightarrow{E_0}[\cos(\omega_0 - \omega_v)t + \cos(\omega_0 + \omega_v)t]$$

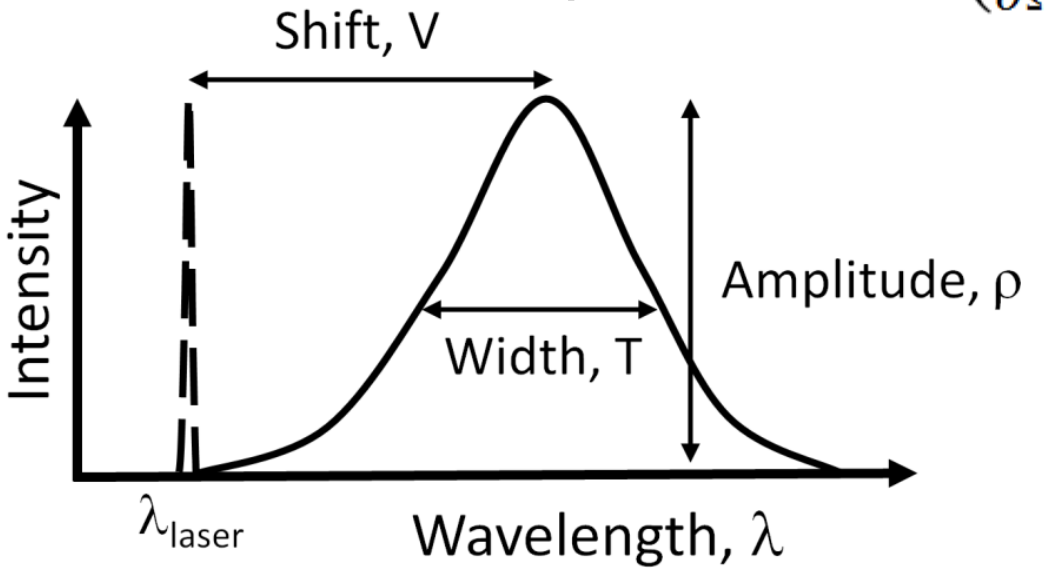
  - Amplitude of Raman is proportional to the magnitude of: $\left(\frac{\partial \alpha}{\partial Q}\right)_0$
  - Polarizability, α, must *change* during vibrations to be Raman active
  - Raman active:
  - Raman inactive:



Rayleigh and Raman Theory 5

- Rayleigh Scattering is at or near the laser's wavelength.
  - Doppler broadened (~$T^{1/2}$)
  - Can be Doppler shifted (~V)
  - Signal proportional to N (~ρ)

$$I_{zz}^{\Omega} = \left(\frac{\partial \sigma}{\partial \Omega}\right)_{zz} NI$$



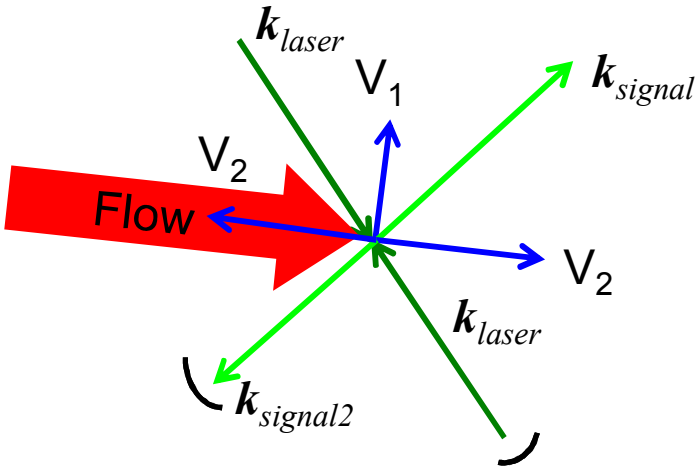
  - Can potentially measure these simultaneously
    - Requires high spectral resolution

Rayleigh and Raman Examples

- Rayleigh
  - Velocimetry, multi point
  - V, T, ρ, single point
  - V, T, ρ, imaging
  - ρ measurement (and infer T)
- Raman
  - Concentration
  - Temperature

Rayleigh Velocimetry

- Rayleigh scattering velocimetry can measure selected velocity components
  - Measured component is bisector between incident laser and collection optics



Wave vector:

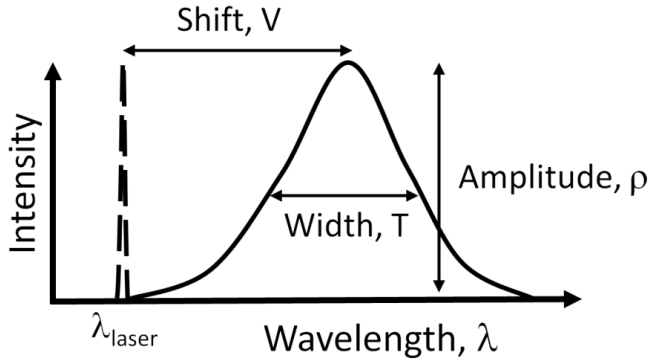
$$k = 2\pi/\lambda$$

D. Bivolaru, P. M. Danehy, R. L. Gaffney, Jr. and A. D. Cutler, "Direct-View Multi-Point Two-Component Interferometric Rayleigh Scattering Velocimeter," 46th AIAA Aerospace Sciences Meeting and Exhibit, Reno, Nevada, AIAA Paper 2008-236, January 7-10, 2008

  - Can measure multiple velocity components by adding additional detection angles or laser beams

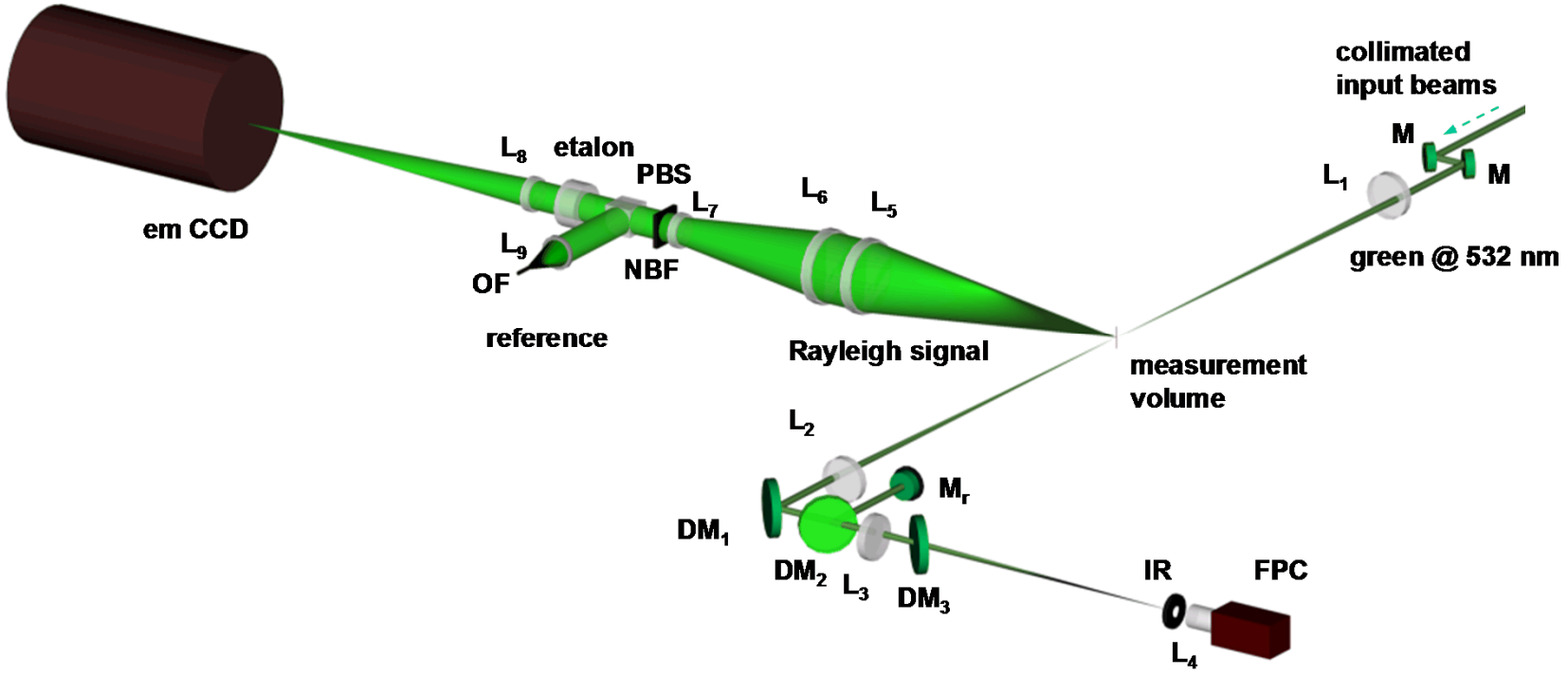
Rayleigh Velocimetry

- Rayleigh scattering velocimetry has been performed different ways, always requiring a high resolution device to resolve the Doppler shift
  - I_2 filter in front of CCD
    - Imaging capability
    - limited dynamic range
  - Fabry Perot etalon
    - PMT based (faster)
    - CCD based (multi-point)
  - Combined I_2 filter and Fabry Perot etalon
  - See manuscript for references



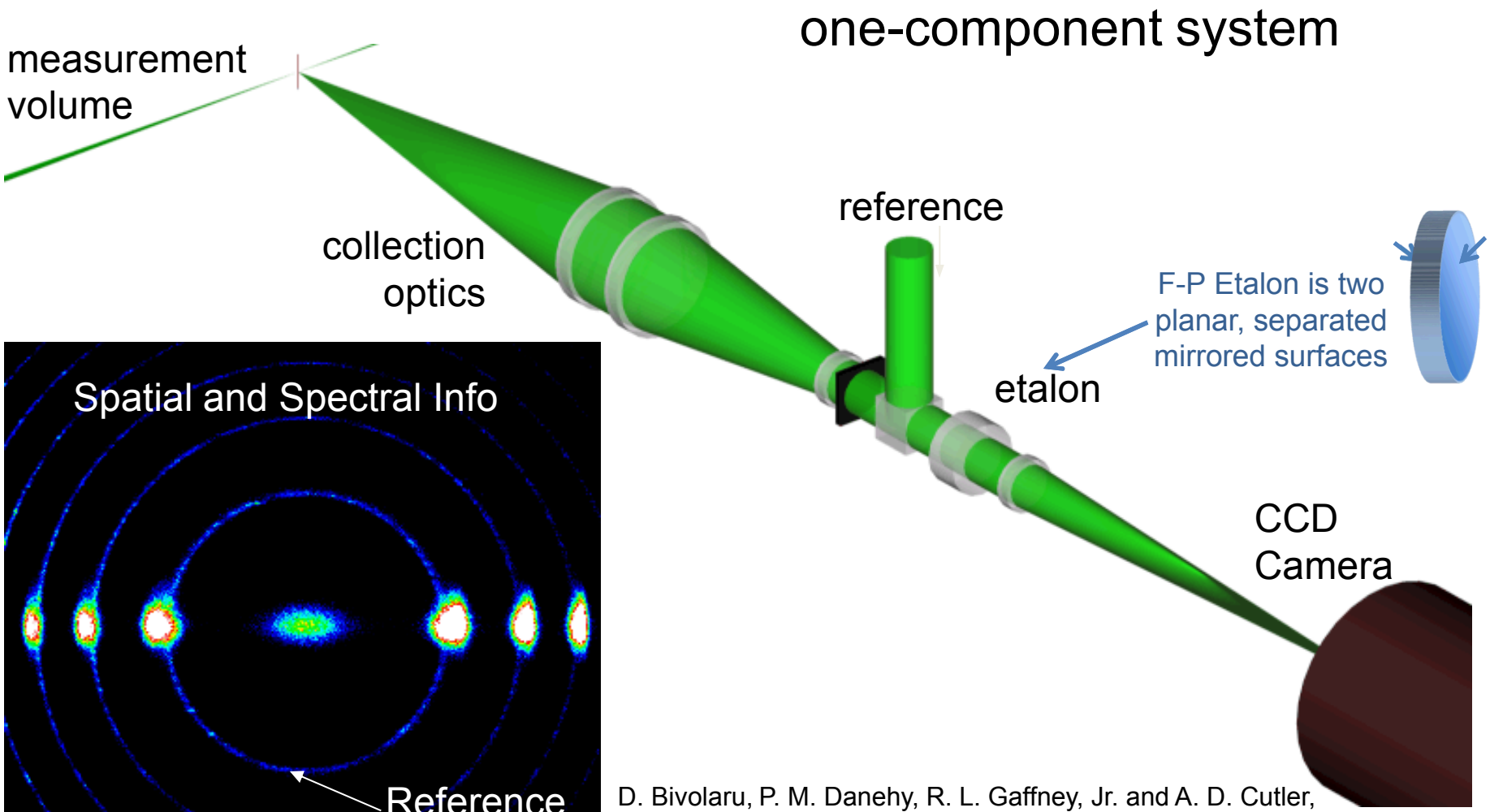
Rayleigh Velocimetry using Fabry-Perot Etalon

- Velocimetry using a Fabry Perot Interferometer (FPI) with CCD camera



- D. Bivolaru, P. M. Danehy, R. L. Gaffney, Jr. and A. D. Cutler, "Direct-View Multi-Point Two-Component Interferometric Rayleigh Scattering Velocimeter," 46th AIAA Aerospace Sciences Meeting and Exhibit, Reno, Nevada, AIAA Paper 2008-236, January 7-10, 2008

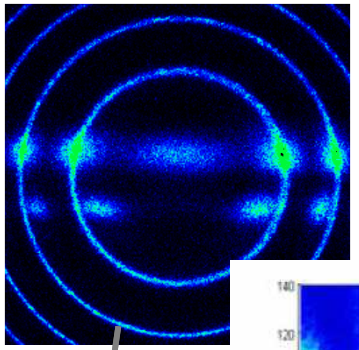
Rayleigh Velocimetry using Fabry-Perot Etalon



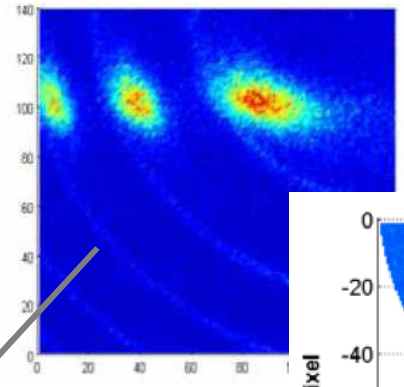
D. Bivolaru, P. M. Danehy, R. L. Gaffney, Jr. and A. D. Cutler, "Direct-View Multi-Point Two-Component Interferometric Rayleigh Scattering Velocimeter," 46th AIAA Aerospace Sciences Meeting and Exhibit, Reno, Nevada, AIAA Paper 2008-236, January 7-10, 2008

Rayleigh Scattering Image Processing

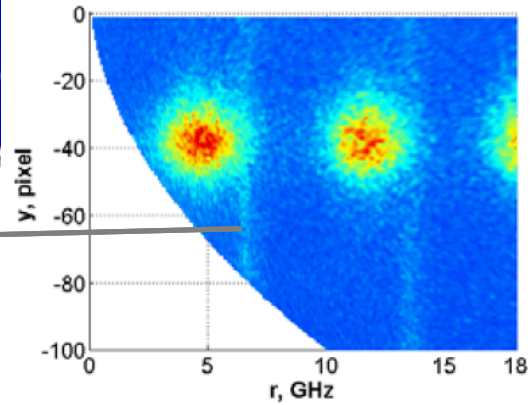
Raw image data



Import to Matlab®

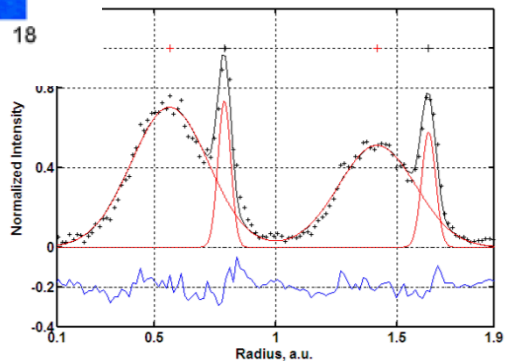


x-*y* to *r*-*y* transform



Circles provide reference
(7.5 GHz etalon)

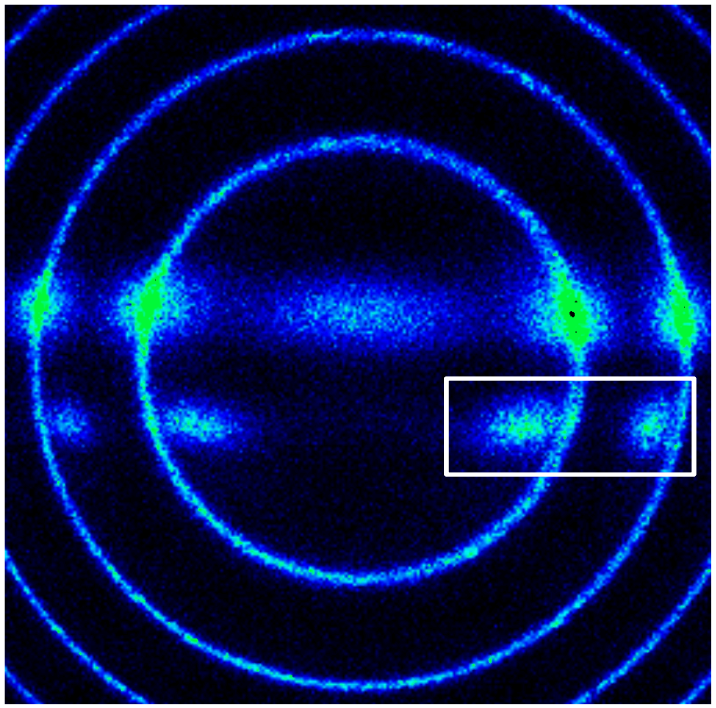
Combine to spectrum and fit

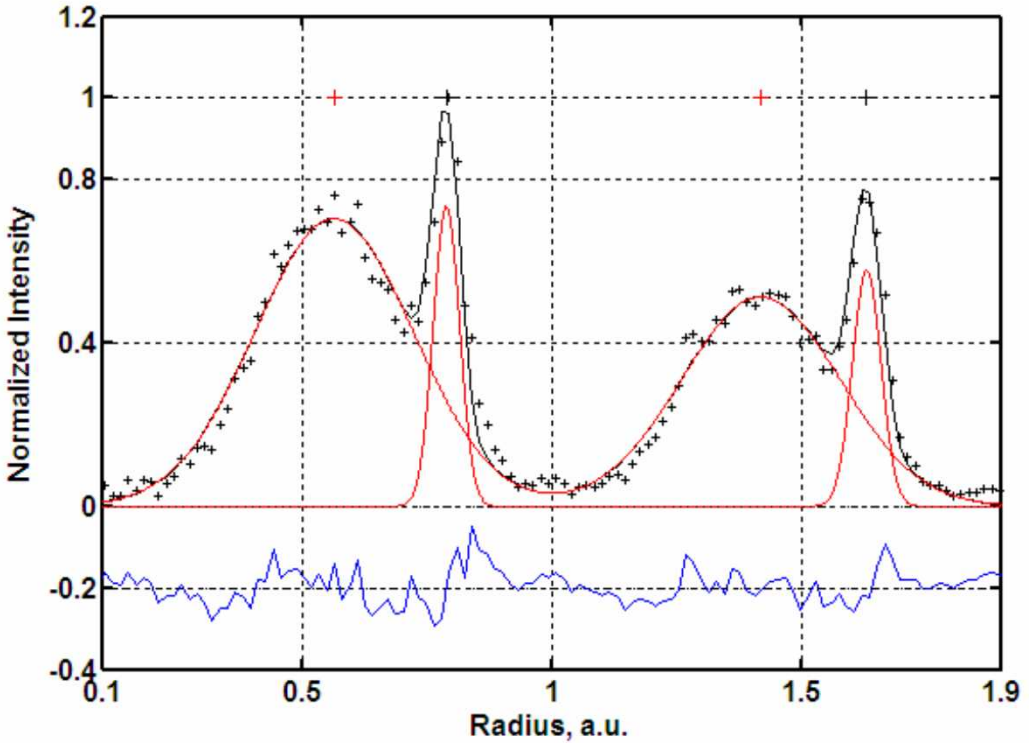


D. Bivolaru, P. M. Danehy, R. L. Gaffney, Jr. and A. D. Cutler, "Direct-View Multi-Point Two-Component Interferometric Rayleigh Scattering Velocimeter," 46th AIAA Aerospace Sciences Meeting and Exhibit, Reno, Nevada, AIAA Paper 2008-236, January 7-10, 2008

- Significant image processing is required

Rayleigh Scattering Velocity Results





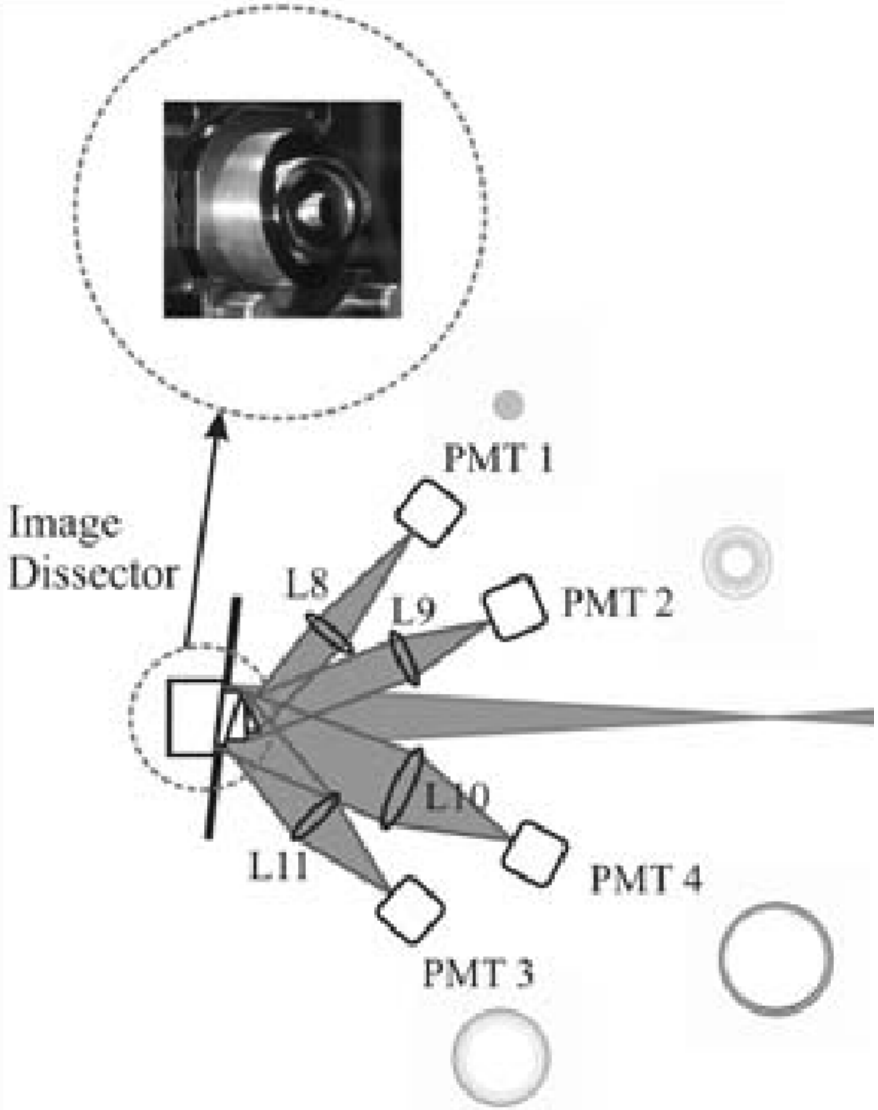
- *Rayleigh scattering in a large scale (6 cm diameter) Mach 1.6 H_2-air combustion heated jet operating at Mach 5.5 enthalpy at NASA Langley*

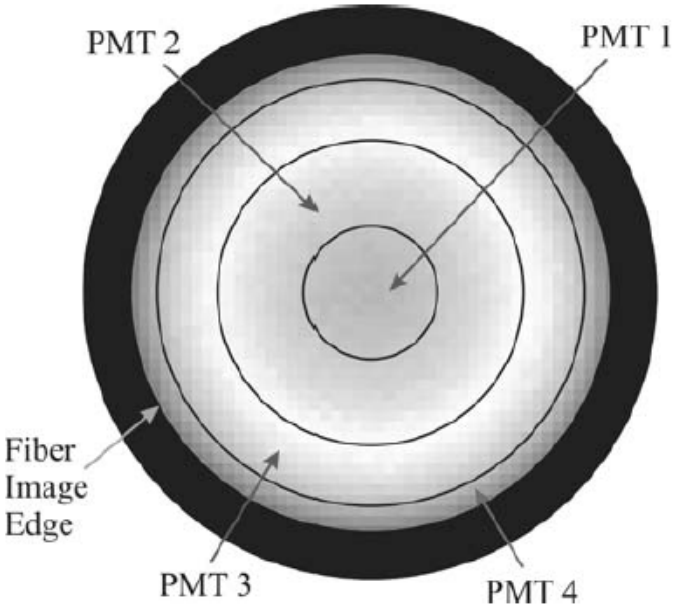
Velocity precision of ~40 m/s out of 1200 m/s, (3%)
The dynamic range of the instrument was ~3000 m/s
- precision 1% of dynamic range

D. Bivolaru, P. M. Danehy, R. L. Gaffney, Jr. and A. D. Cutler, "Direct-View Multi-Point Two-Component Interferometric Rayleigh Scattering Velocimeter," 46th AIAA Aerospace Sciences Meeting and Exhibit, Reno, Nevada, AIAA Paper 2008-236, January 7-10, 2008

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Rayleigh Scattering: T, V, ρ

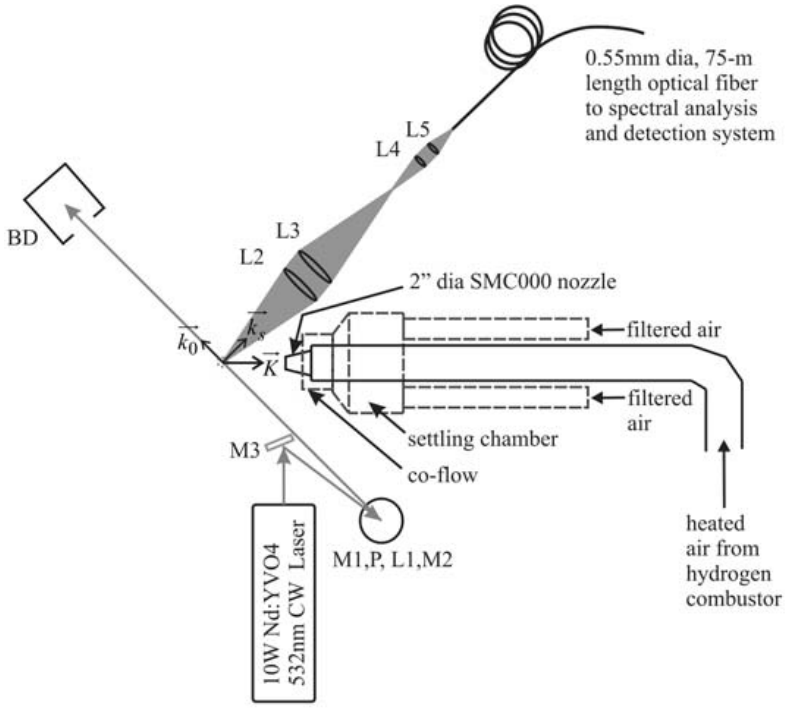


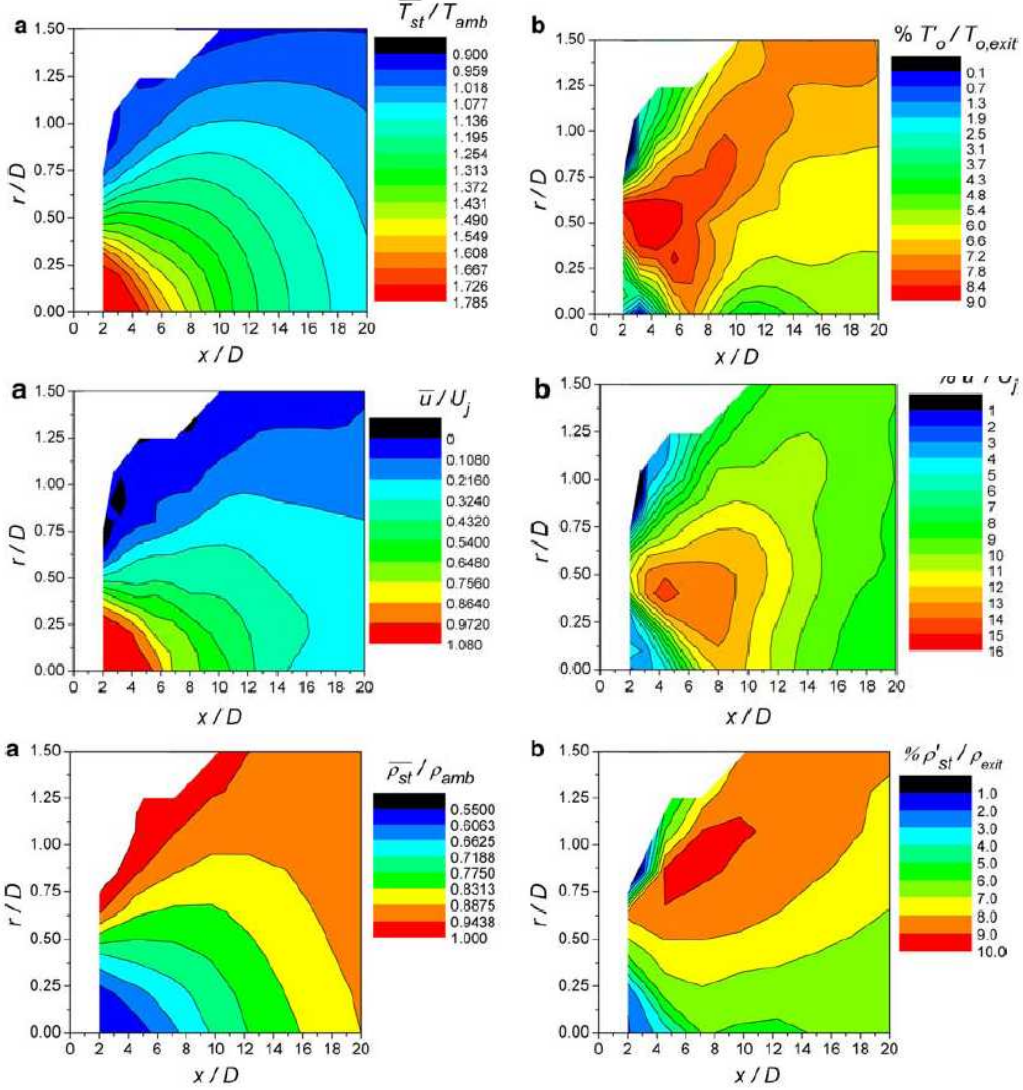


- Continuous laser excitation
  - 10 W at 532 nm
- Fiber optic collection
- Data rate: 10 kHz
- Single point measurement

A. F. Mielke and K. A. Elam. "Dynamic measurement of temperature, velocity, and density in hot jets using Rayleigh scattering." *Experiments in fluids* 47, no. 4-5 (2009): 673-688.

Rayleigh Scattering: T, V, ρ



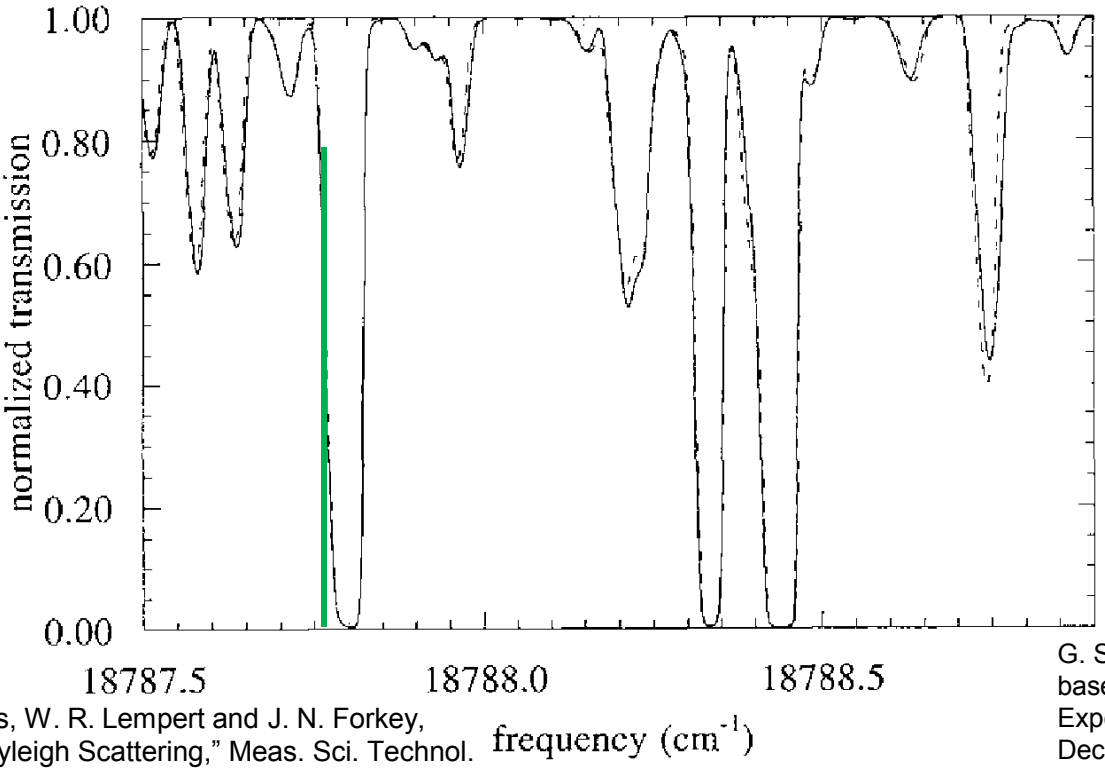


- Heated, 2” (50 mm) diameter axisymmetric near-sonic jet at NASA Glenn Research Center

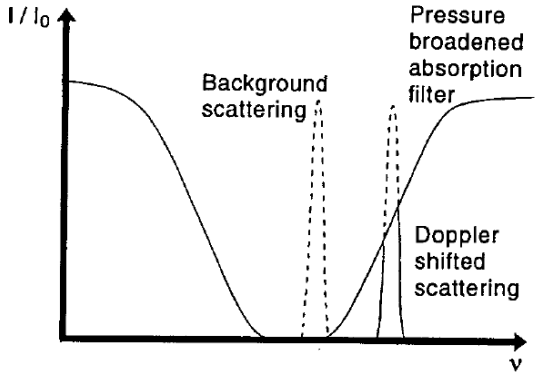
A. F. Mielke and K. A. Elam. "Dynamic measurement of temperature, velocity, and density in hot jets using Rayleigh scattering." *Experiments in fluids* 47, no. 4-5 (2009): 673-688.

Rayleigh Velocimetry (I_2 filter)

- Instead of an etalon, use I_2 filter → 2D imaging



I_2 absorption near 532 nm



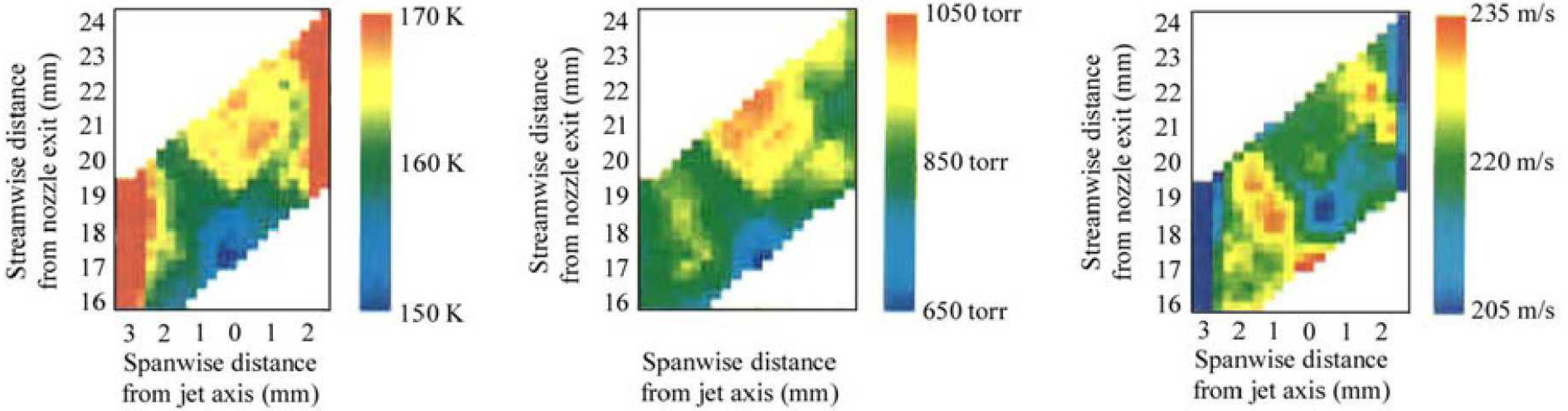
R. B. Miles, W. R. Lempert and J. N. Forkey, "Laser Rayleigh Scattering," Meas. Sci. Technol. p. R33-R51, v.12, 2001

G. S. Elliott, M. Samimy, S. A. Arnette "A molecular filter based velocimetry technique for high speed flows," Experiments in Fluids, Volume 18, Issue 1-2, pp 107-118, December 1994

- Gas cell, filled with low-pressure N_2 plus some I_2 crystals; control temperature to determine absorption
- Scan Nd:YAG laser frequency (or fixed)

Rayleigh Imaging of T, V, ρ

- Direct YAG laser sheet into a Mach 2 jet

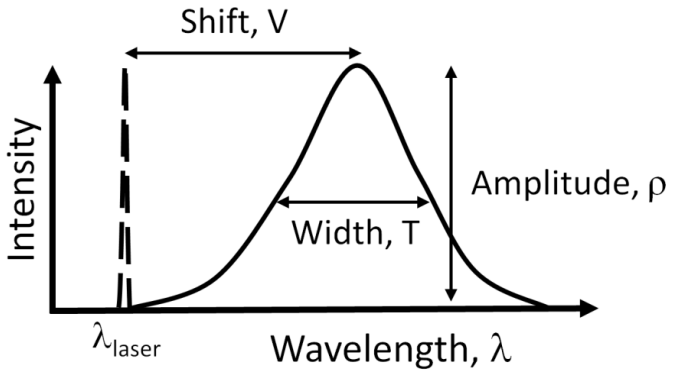


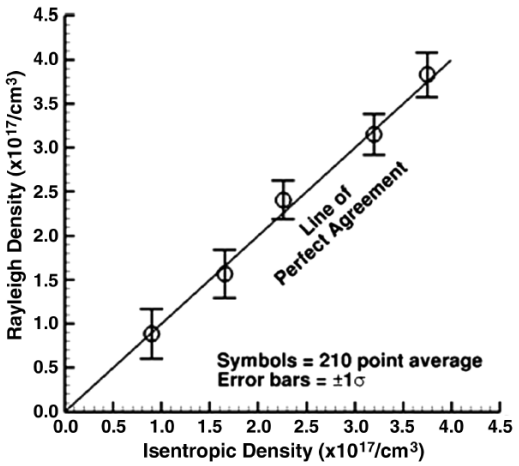
R. B. Miles, W. R. Lempert and J. N. Forkey, "Laser Rayleigh Scattering," Meas. Sci. Technol. p. R33-R51, v.12, 2001

- Scanned laser frequency, determine T, V, ρ (report P) based on measured spectrum at each pixel
- Same method can be used for velocimetry from particles (usually fixed frequency, multiple detection angles)
  - Planar Doppler Velocimetry (PDV) or Doppler Global Velocimetry (DGV)

Rayleigh Scattering: ρ

- Rayleigh scattering has a spectrum:
  - Sensitive to T, V, ρ
  - Prior examples used Fabry Perot Etalon or I_2 cell to observe spectrum
  - These devices reject signal, making them less sensitive detectors of ρ.
- Collect *all* of the scattering
  - Less equipment/less setup/simpler
  - Bigger signal
  - Can do single point or imaging
  - Only sensitive to ρ.
    - (If you know P and composition you can convert ρ to T.)



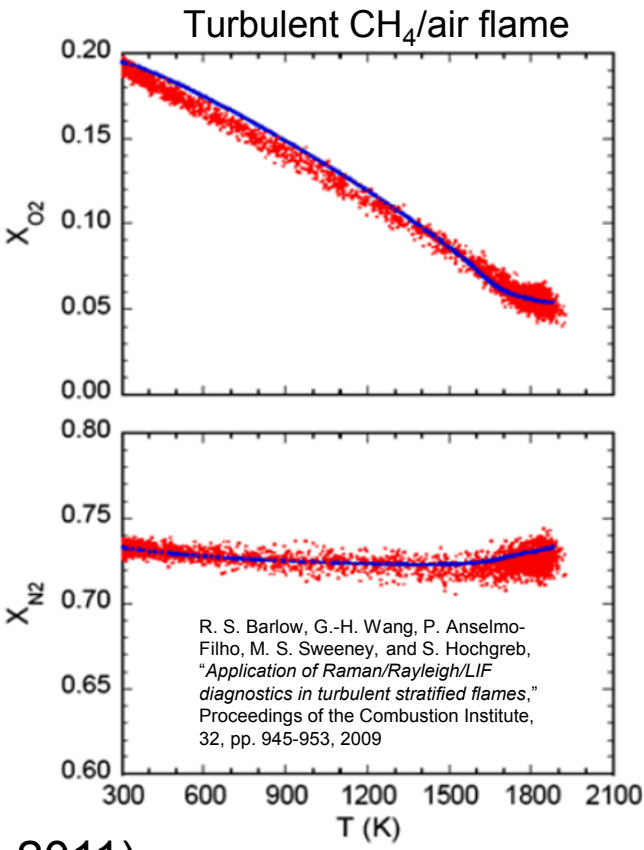


Using a UV laser (e.g. ArF at 193 nm) allows measurements to be made at low pressures (e.g. 1/300 atmospheric density)

Balla, R. Jeffrey, and Joel L. Everhart. "Rayleigh scattering density measurements, cluster theory, and nucleation calculations at Mach 10." AIAA Journal 50, no. 3 (2012): 698-707.

Rayleigh Scattering: ρ

- When precise measurements are required, high S/N is needed
  - Use high powered lasers, focused to a point
    - If too high: laser-induced breakdown
- Sandia, Livermore, Calif.:
  - Raman/Rayleigh/LIF line imaging at P = 1 atm
  - 4 double-pulse Nd:YAG lasers: 1.8 J/pulse
  - Camera images Rayleigh along line
    - ~10 pix / mm along 6-mm-long probe volume
  - N_2, O_2, CH_4, CO_2, H_2O and H_2 from Raman
  - CO from 2 photon LIF
  - Temperature from Rayleigh density, ρ,
    - Need composition to determine $\left(\frac{\partial \alpha}{\partial Q}\right)_0$
    - If known P, idea gas law give T
    - T precision reported: ~0.75%
    - (ρ precision is probably same)
  - Cutting edge MHz laser: 40x lower energy (Jiang, 2011)



Raman Spectroscopy

- Raman spectrum obtained in a CH_4-air flame:
  - Average of 500 pulses using stretched 0.5 J/pulse, 532 nm
    - Stretch 10 ns pulse to 500 ns to avoid laser-induced breakdown

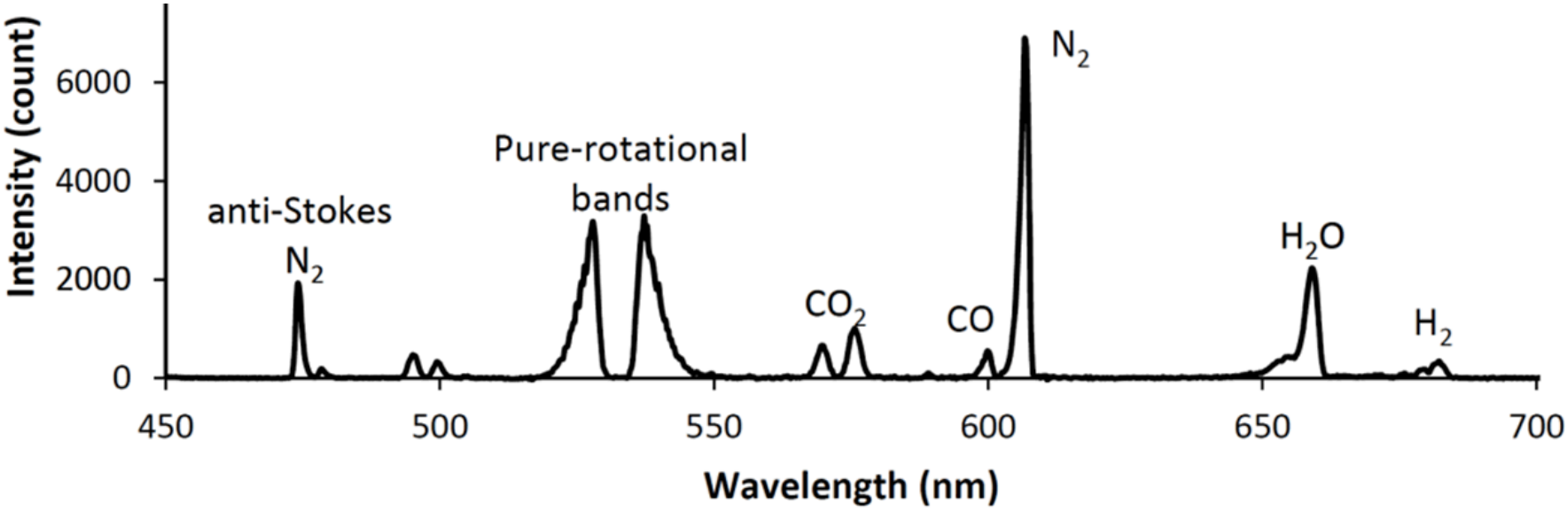


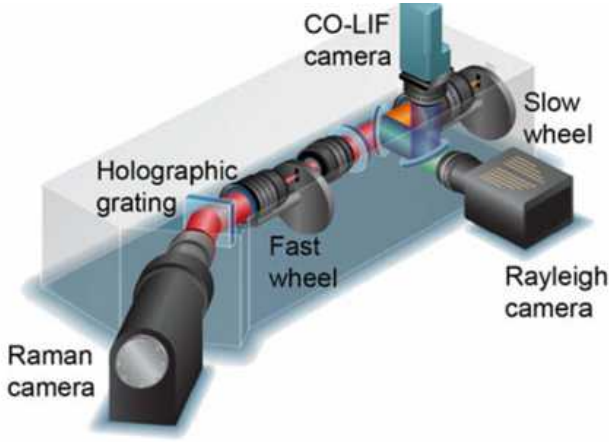
Figure courtesy of Jun Kojima, NASA Glenn Research Center (Jun.J.Kojima@nasa.gov)

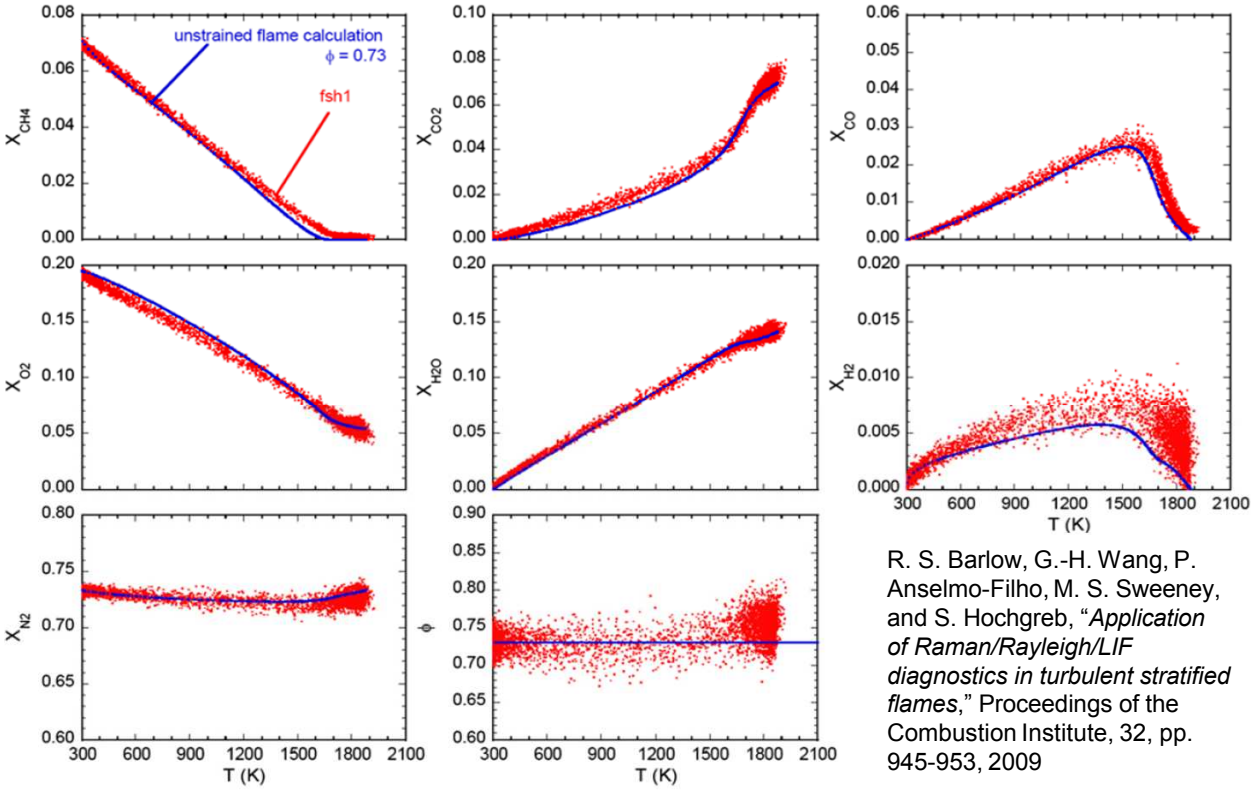
- Can measure species (N_2, CO, CO_2, H_2, H_2O) from such spectra &temperature from Stokes/anti-Stokes

Raman Concentration Measurement

(and Rayleigh Temperature and LIF concentration)

- Example of Raman concentration measurements in a subsonic turbulent CH_4/air flame at Sandia National Laboratories, California
  - 4 Nd:YAG lasers at 532 nm to produce 1.8 Joules/pulse, focused to a point





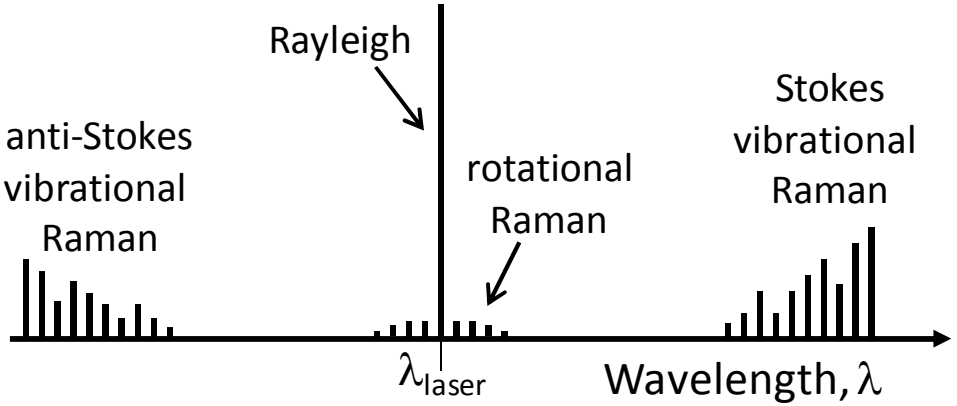
χ_i precision ~0.7% to 7.5%
T precision ~0.75%

R. S. Barlow, G.-H. Wang, P. Anselmo-Filho, M. S. Sweeney, and S. Hochgreb, "*Application of Raman/Rayleigh/LIF diagnostics in turbulent stratified flames*," Proceedings of the Combustion Institute, 32, pp. 945-953, 2009

- Raman/Rayleigh/LIF line imaging (~10 pixels / mm along the 6-mm-long probe volume)
  - N_2, O_2, CH_4, CO_2, H_2O and H_2 from Raman, CO from 2 photon LIF
  - Temperature from Rayleigh density measurement (idea gas law)

Raman Temperature Measurement

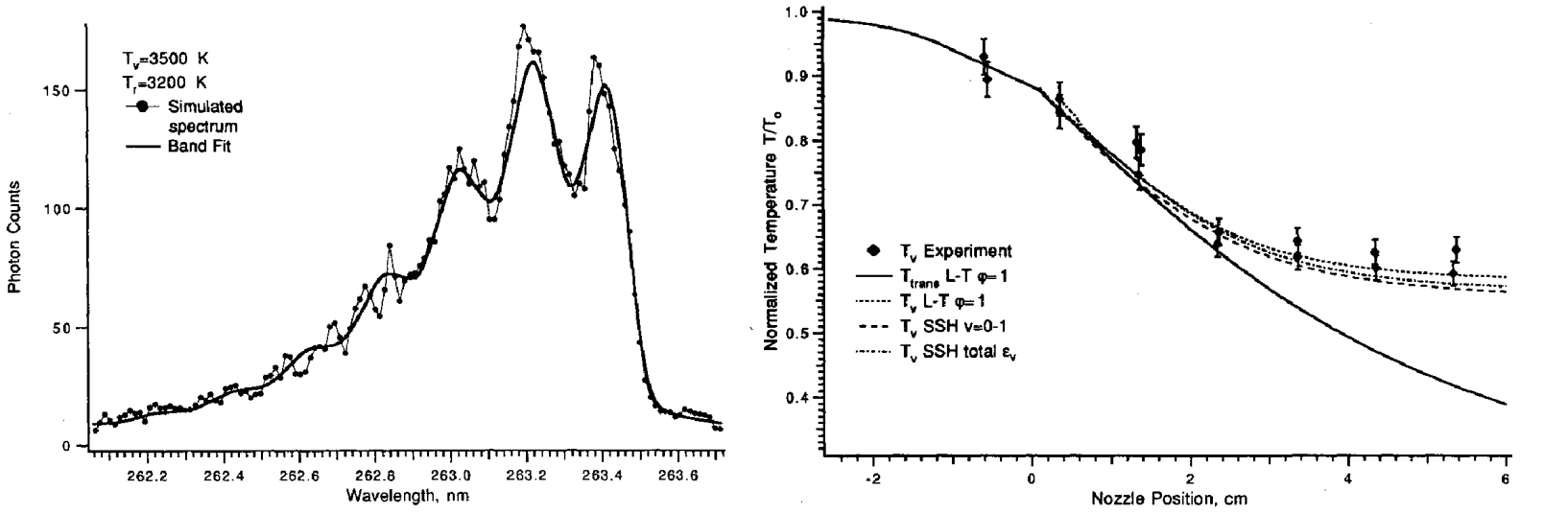
- Several ways to measure temperature with Raman
  - none as precise as CARS (3%)



- Rotational Temperature from pure-rotational Raman
- Vibrational temperature measurements (see Kojima, 2008)
  - Only good above 700 K
  - Stokes / anti-Stokes vibrational Raman
    - Low resolution, so can measure concentrations at same time
    - ~20% precision in measurements along a line (Wehrmeyer, 1996)
  - Spectrally resolved vibrational band (Sharma et al, 1993)

Raman Temperature Measurement

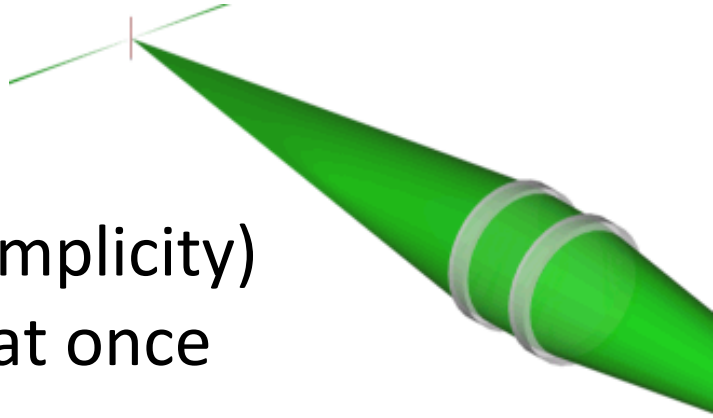
- Raman temperature measurement from spectrally resolved vibrational N_2 band.
  - Excited with KrF Excimer laser 0.25 J at 248 nm (25 ns pulse)



S. P. Sharma, S. M. Ruffin, W. D. Gillespie and S. A. Meyer, "Vibrational Relaxation Measurements in an Expanding Flow Using Spontaneous Raman Scattering," J. of Thermophys. and Heat Trans. Vol. 7, No. 4, Oct.-Dec. 1993

  - Single shot precision ~5% (not explicitly stated)
    - More precise than Stokes/anti-Stokes but cannot measure species concentrations with same instrument.

Advantages and Disadvantages: Rayleigh & Raman



- Advantages:
  - No seeding required
  - Only one color of laser used (simplicity)
  - Multiple properties measured at once
    - T, V, ρ for Rayleigh
    - T, many species for Raman
    - Can do Raman and Rayleigh together and with LIF
- Disadvantages:
  - Weak signal:
    - Requires very high power laser
      - stretched to avoid laser induced breakdown
    - Requires large windows, big, close collection optics
    - Usually singe point or line measurements
  - Temperature less accurate & precise than CARS

Coherent anti-Stokes Raman Spectroscopy (CARS)

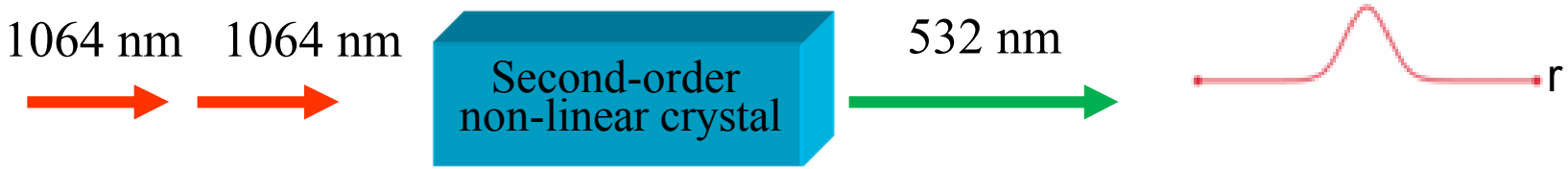
Nonlinear Optics

- First, a few words about nonlinear optics
  - Conservation of energy
  - Conservation of momentum
  - Wave equation from Maxwell's equations

- Second Harmonic Generation as an Example
  - 2^{nd} order nonlinearity (solid)
  - CARS is a 3^{rd} order nonlinearity (gas)

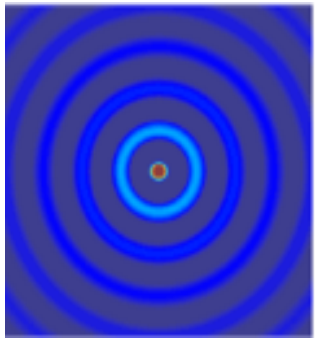
Intro: Nonlinear Optics

– **Second Harmonic Generation (SHG)**



– Solved using the Wave Eqn:

$$\nabla^2\mathbf{E} - \frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} = \mu_0\frac{\partial^2\mathbf{P}_{NL}}{\partial t^2}$$

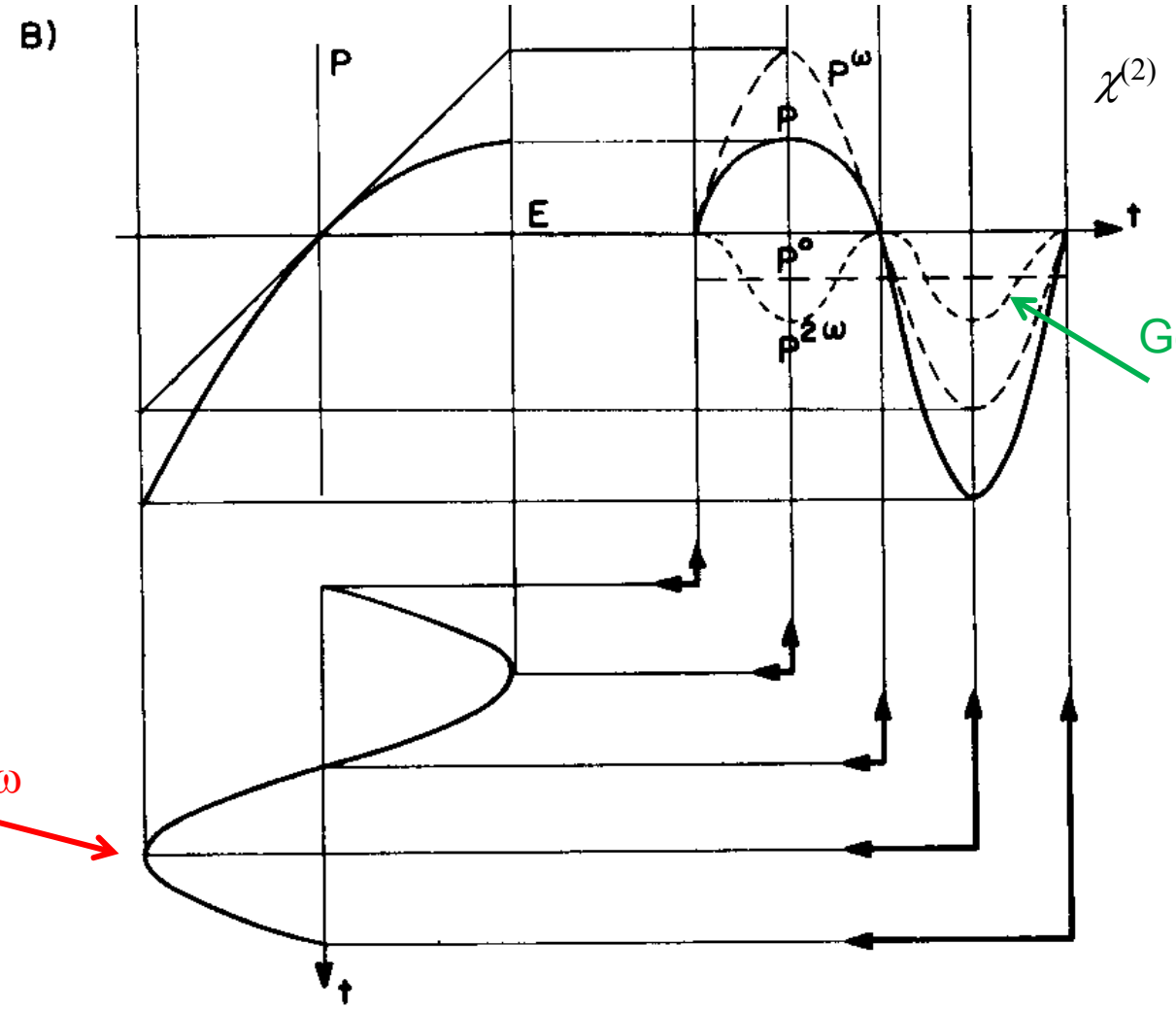


– Assume SVEA and 1st Born Approx:

$$I_{\omega_2} \propto \frac{d^2 P_{\omega_1}^2 L^2}{A^2}$$

– d is a coefficient, P is power, L is length, A is area

Non-linear Susceptibility

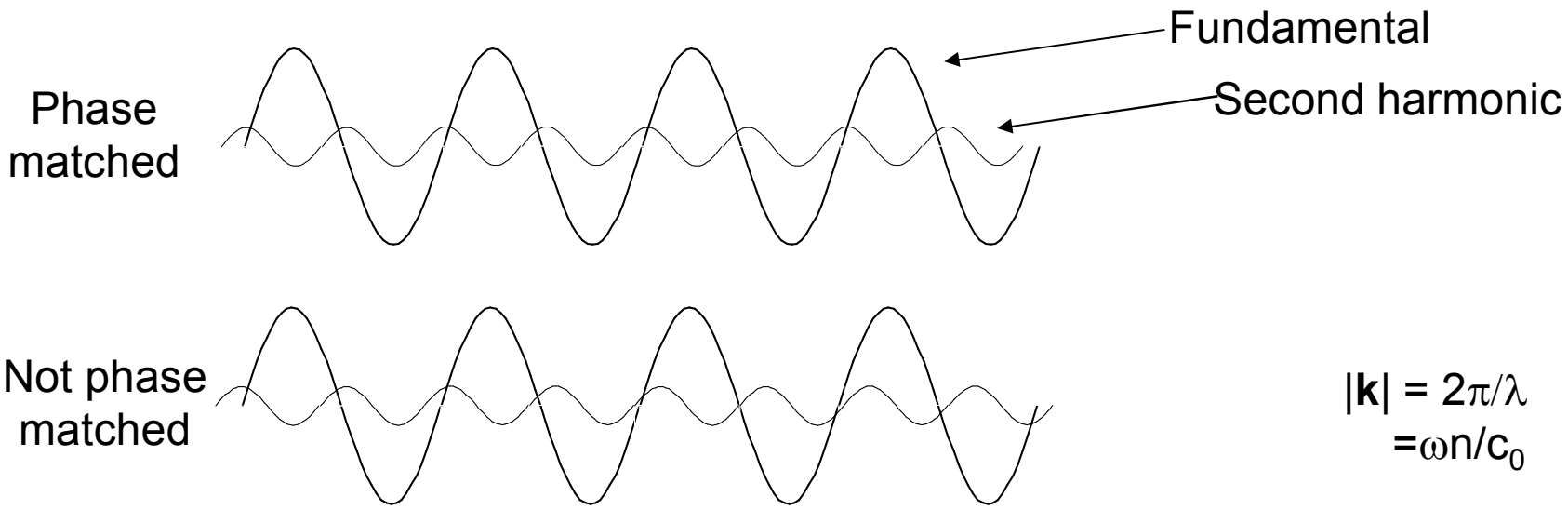


G.C. Baldwin "An introduction to nonlinear optics." *New York: Plenum Press,* (1969).

- Non-linear effects: Intense electric fields of one frequency can generate a **P** at another

Phase Matching

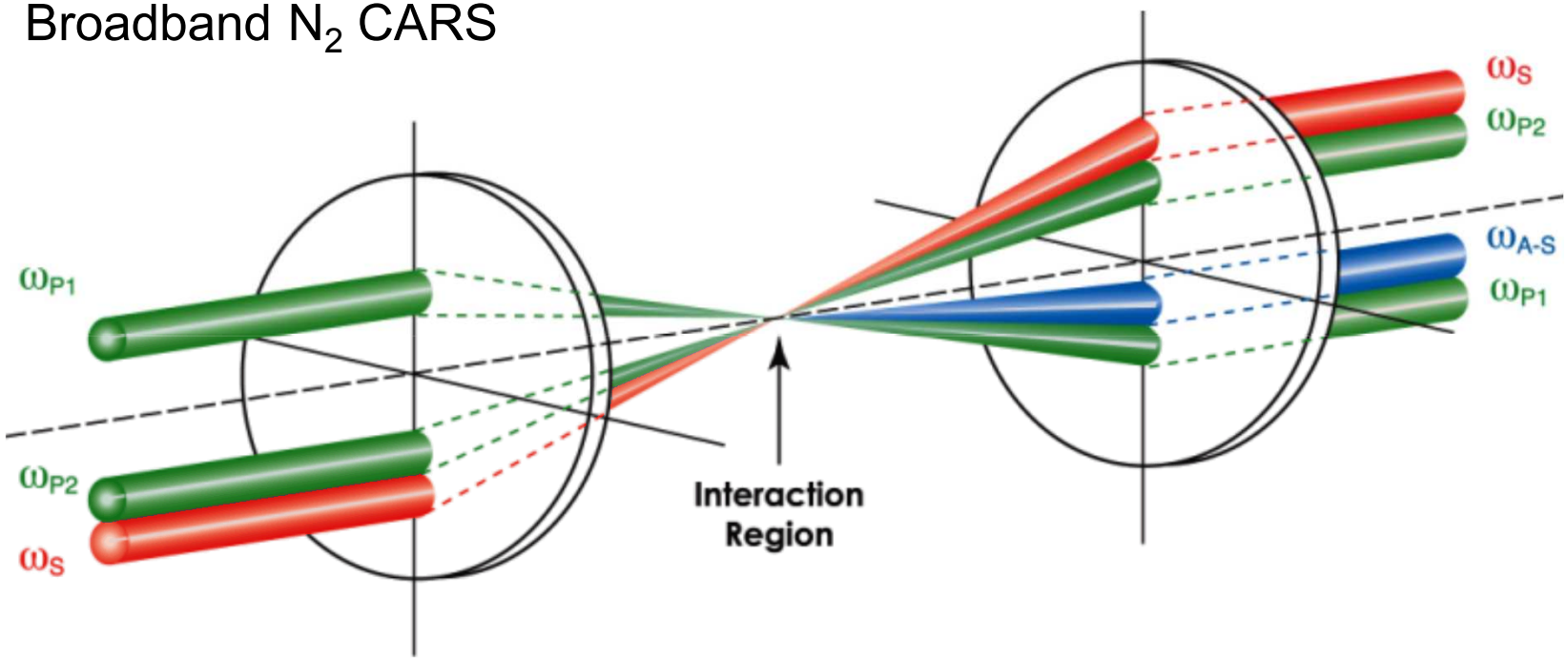
- What is Phase Matching?
  - *Ensuring that the light wavefronts 'match up'.*



  - Dispersion causes deconstructive interference
  - New photons generated are out of phase with previous ones: resulting in loss
  - For SHG there are tricks to make it phase matched
- Conservation of momentum: phase matching
- Conservation of energy: $\omega_2 = 2\omega_1$

What is CARS?

Broadband N_2 CARS



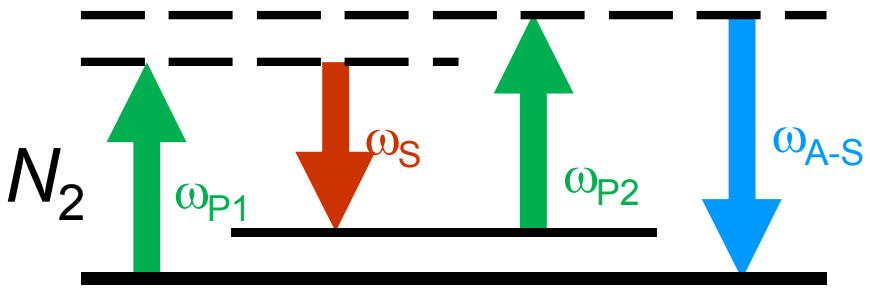
Spectrally-broad red beam +

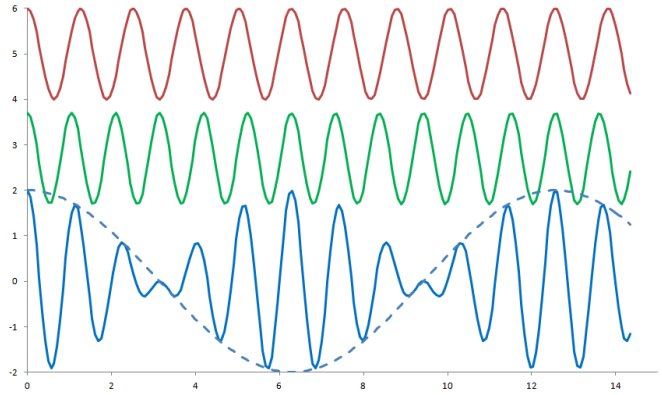
Spectrally-narrow green beams =

Single-shot blue CARS spectrum

CARS Energy Level Diagram (conservation of energy)

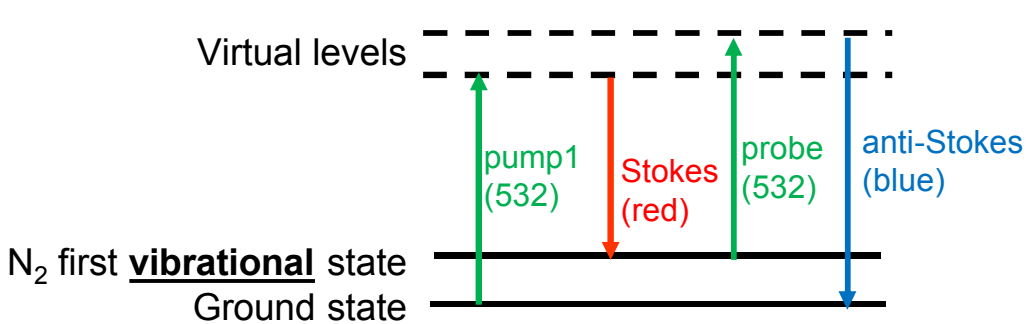
- Lasers: 2 Green (532 nm) and 1 Red (607 nm)
  - Frequency difference between Green and Red Probes N_2 vibrational Raman Shift



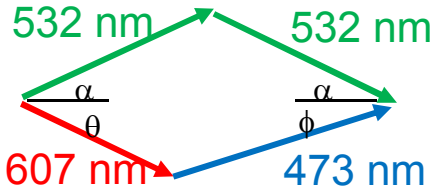


- Result: Blue signal beam at 473 nm.
  - Spatially, spectrally, temporally filter this signal
    - Good technique for luminous flows
  - Send to spectrometer equipped with CCD camera
  - Measured CARS spectra is best fit to a library of theoretical spectra on a single-shot basis (10 ns)

CARS Phase Matching (conservation of momentum)



Planar Boxcars Geometry



$$k_{p1} + k_{p2} = k_S + k_{A-S}$$

It is straight forward to compute the CARS phase matching angles:

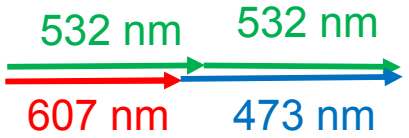
N2 cars		adjust inpu	
	lambda	wavenum	angle (degrees
stokes	607	16474.46	**3.963**
pump 2	532	18796.99	**3.500**
pump 1	532	18796.99	**3.500**
anti stokes	473.4956	21119.52	**3.091**
			0.023576939
N2 Raman =	2322.528	cm-1	0.023576939
(nominal N2 = 2330 cm-1)			*3.00927E-20*

Different molecules require different angles (can get multiple signals at different spatial locations: Dual-Stokes)

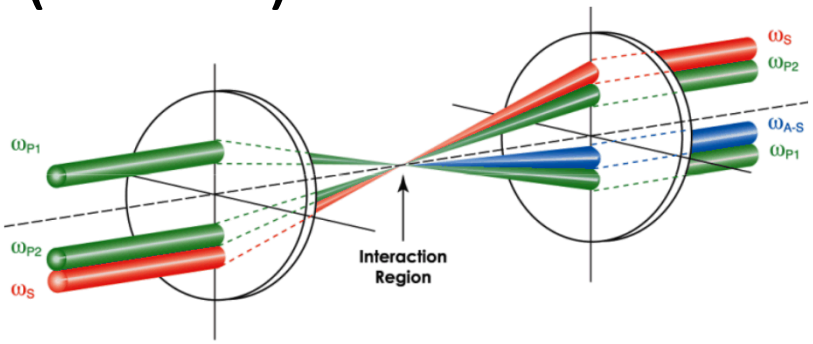
CARS Phase Matching Geometries

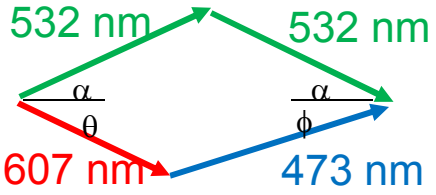
- Collinear CARS:
  - Poor spatial resolution

$$k_{p1} + k_{p2} = k_S + k_{A-S}$$

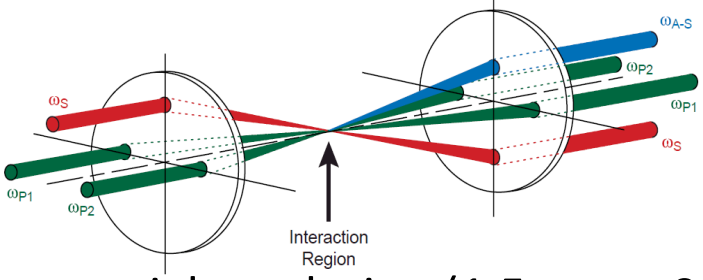


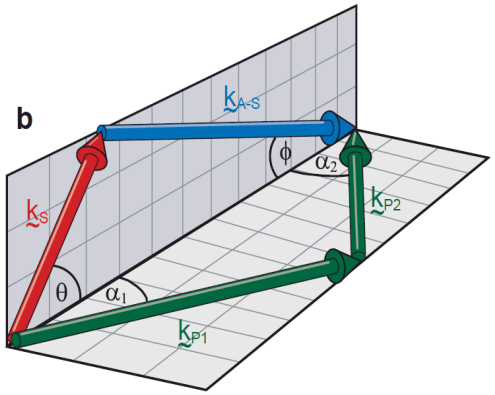
- (Planar) BoxCARS:





- Folded BoxCARS:

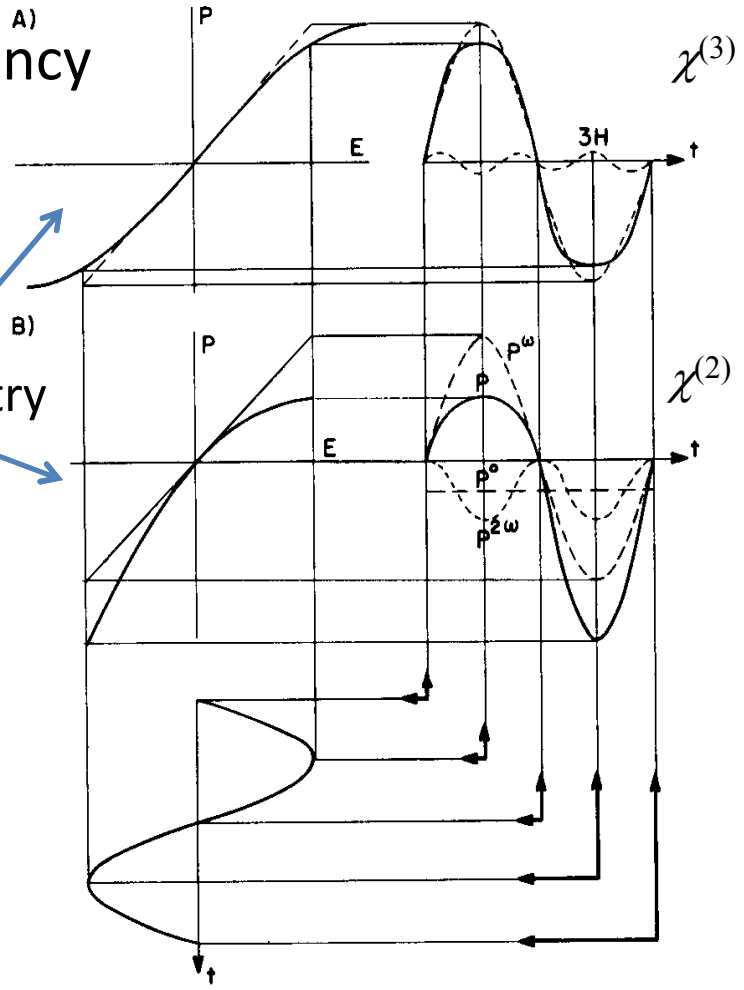




- Best spatial resolution (1.5 mm x 0.1 mm x 0.1 mm) but lower signals and affected by beam steering from turbulence

CARS Theory 1

- Non-linear effects:
  - Intense electric fields of one frequency can generate a P at another
  - $P = \varepsilon_0 (\chi E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots)$
  - $\chi^{(2)}$ = 2nd order susceptibility
    - Only media lacking a centre of symmetry (ie, no gases, liquids)
  - $\chi^{(3)}$ = 3rd order susceptibility
    - All dielectric media
  - Generally ($\chi E >> \chi^{(2)} E^2 >> \chi^{(3)} E^3$)



G.C. Baldwin "An introduction to nonlinear optics." *New York: Plenum Press*, (1969).

CARS Theory 2

- Polarization generated at CARS signal frequency:

$$P^{(3)}(\omega_{signal}) = \epsilon_0 \chi_{CARS} E(\omega_{pump}) E(\omega_{Stokes}) E(\omega_{probe})$$

- Substitute into the wave equation. Result:

$$I_{signal} \propto I_{pump} I_{Stokes} I_{probe} |\chi_{CARS}|^2 L^2$$

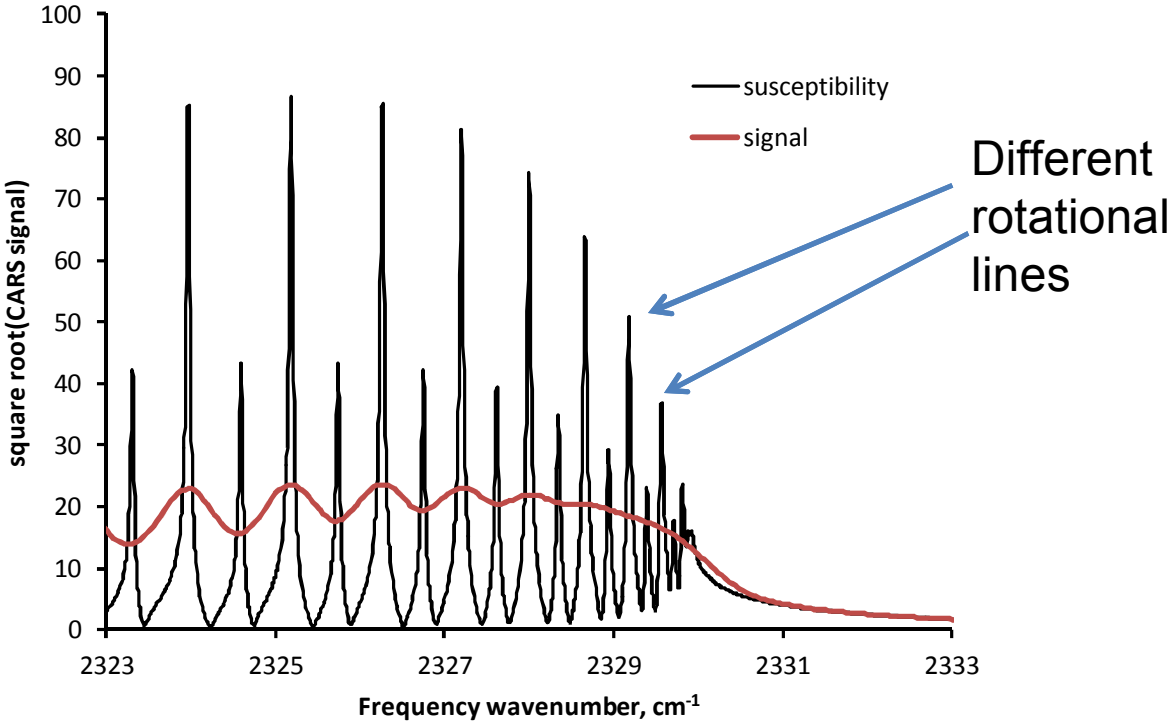
- To obtain large signals, want long L, high laser power (tight focus) and strong resonance

$$\chi_{CARS} = \sum_j \frac{K_j \Gamma_j}{2\Delta\omega_j - i\Gamma_j} + \chi_{nr} \text{ where } K_j \propto N\Delta_j \left(\frac{\partial\sigma}{\partial\Omega}\right)_j$$

- Γ_j is damping coefficient, N is number density, Δ_j is the population difference
  - N→ concentration; Δ_j→Temperature sensitivity

CARS Theory 3

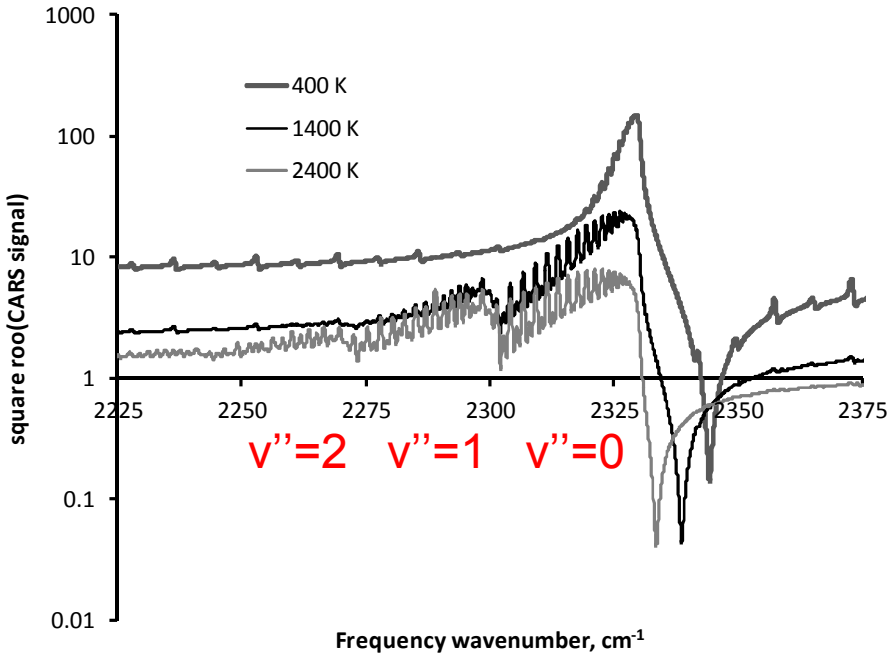
- Instructive to graph example N_2 spectra:



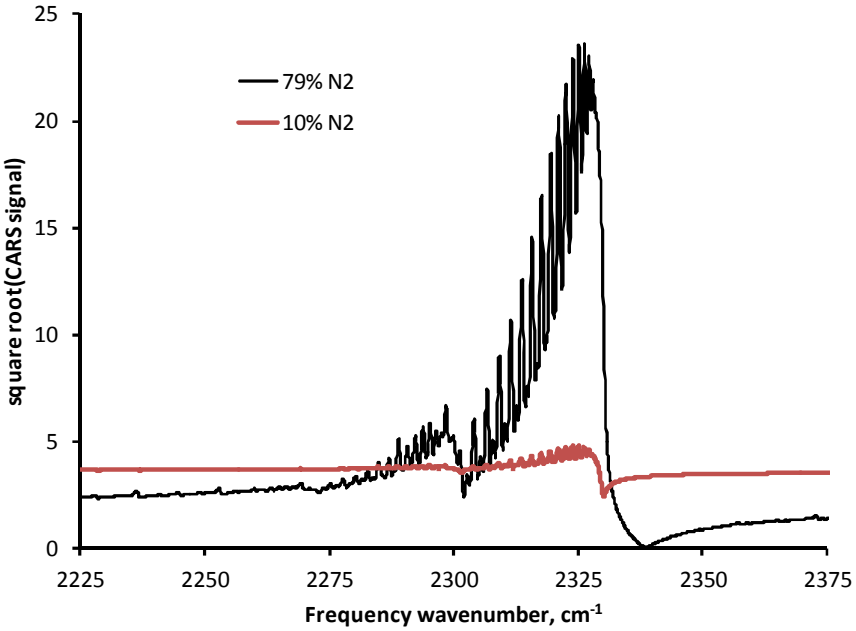
- N_2 is commonly probed with CARS because it is often present and spectroscopy is well known
- CARS spectrum at 1 atm., 1400 K, v" = 0 band.
  - N_2 Q-branch ($\Delta v=+1$, $\Delta J=0$)
  - CARS signal spectrum is convolved with lasers used

CARS Theory 4

- Effects of temperature (left) and composition (right):



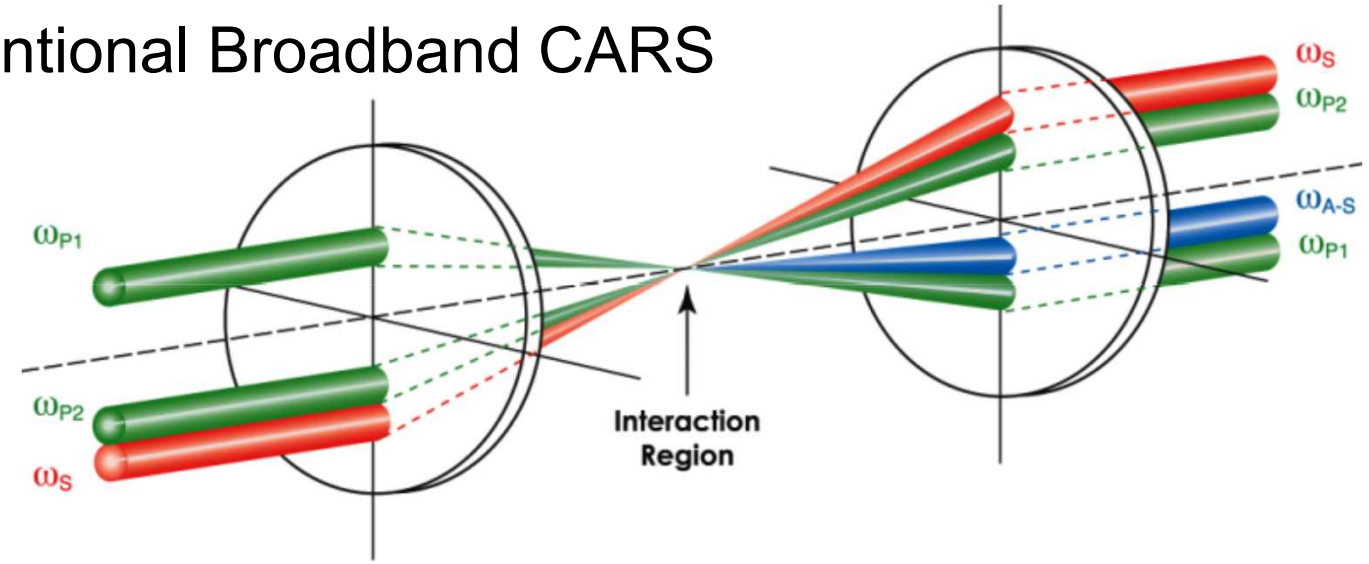
Vibrational "hot band" becomes populated above 700 K



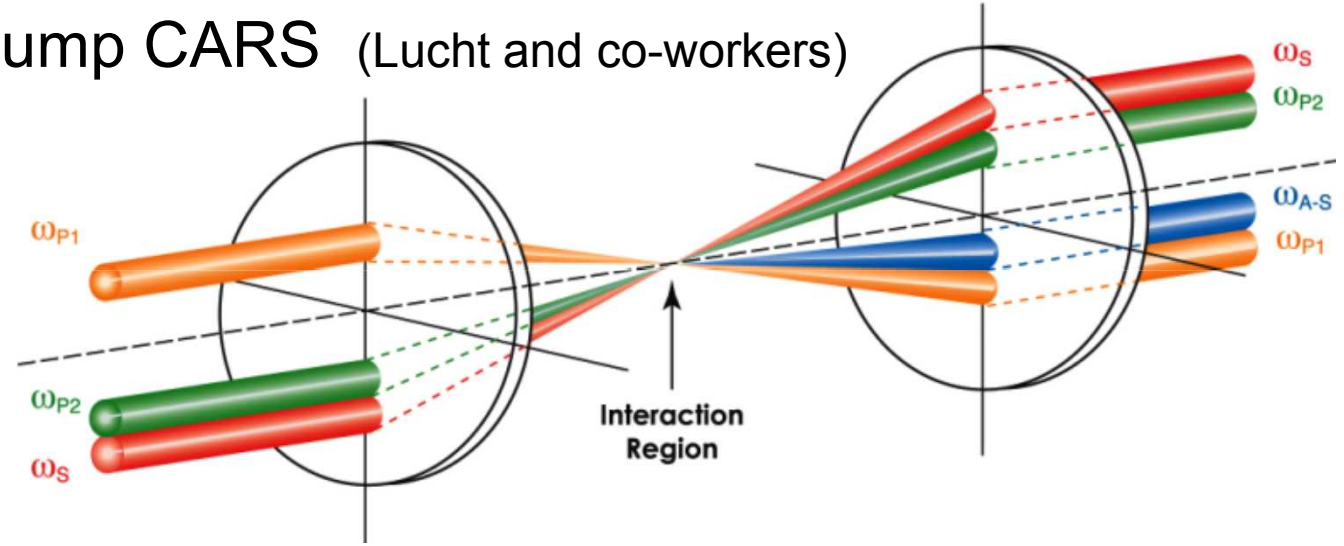
CARS spectra are sensitive to the gas composition through resonant and nonresonant parts

Multi-Species Measurement: Dual-Pump CARS

Conventional Broadband CARS

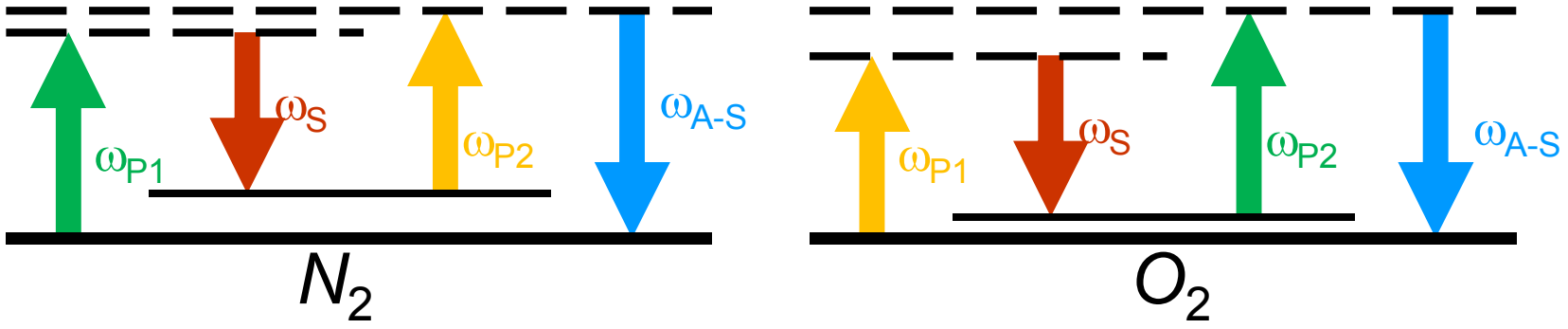


Dual-Pump CARS (Lucht and co-workers)



Dual-Pump CARS: N_2, O_2, H_2

- Green and Red Probe N_2 Raman Shift
- Red and Yellow Probe O_2 Raman Shift

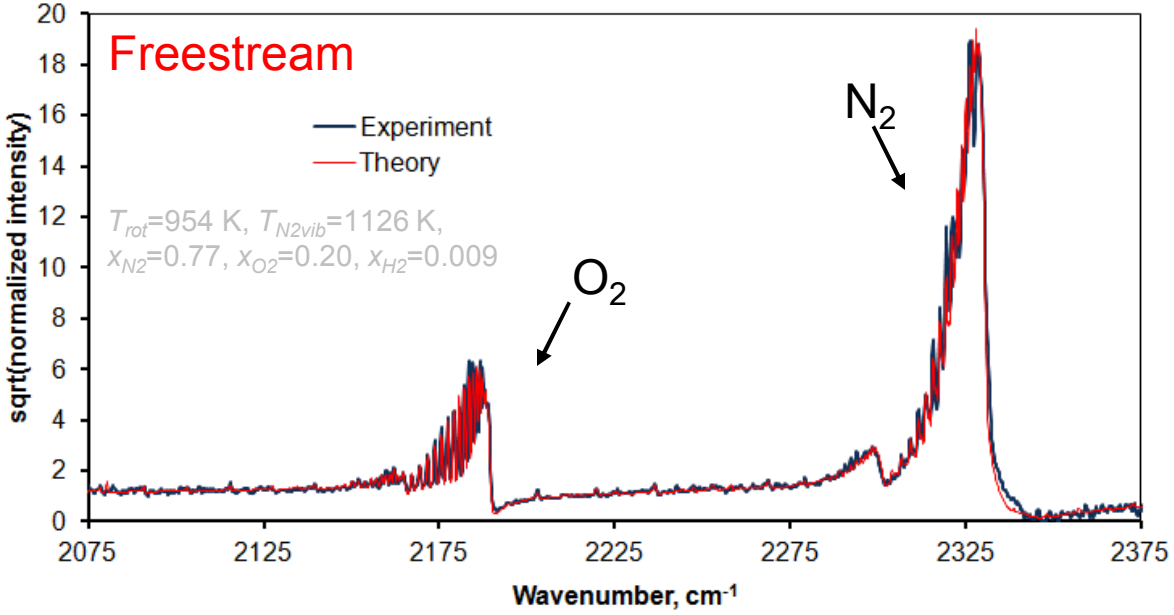


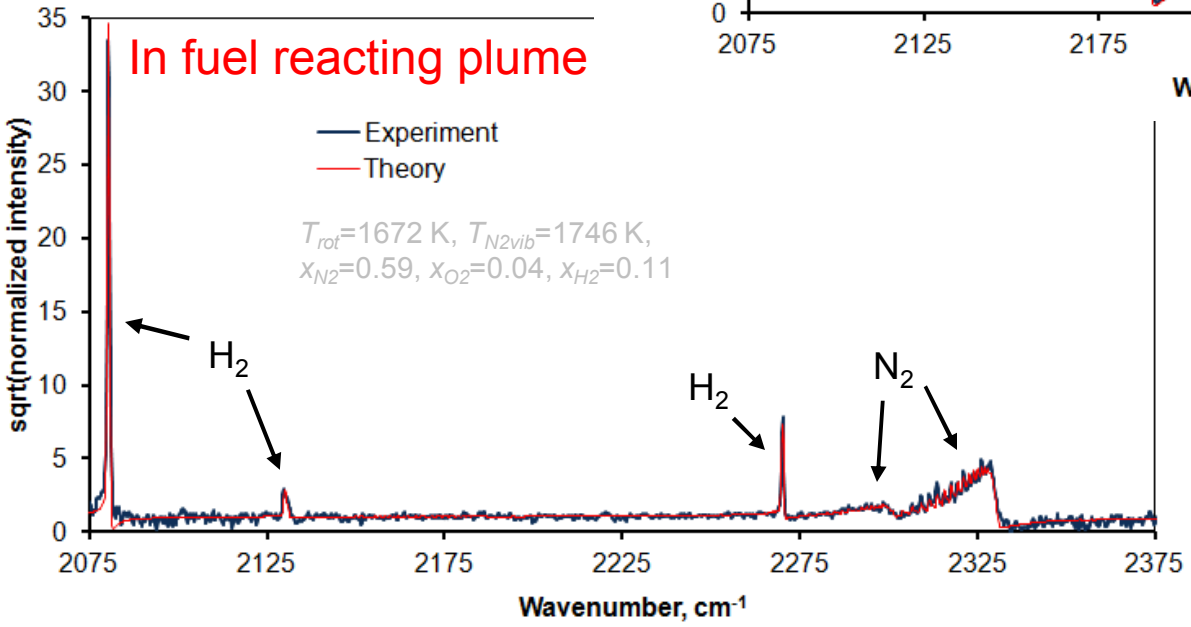
- Result: Two different spectral regions of Raman Shift appear coincident in the 490 nm CARS beam.
  - 'self calibrating' compared to other methods
  - H_2 pure rotational Raman lines present

O'Byrne et al AIAA Journal v. 45, 2007

Typical Dual-Pump CARS Spectra and Fits

- Single-shot spectra obtained in combustor shown with best fit
- Absolute measurement of rotational, vibrational temperature, N_2, O_2, and H_2 mole fractions

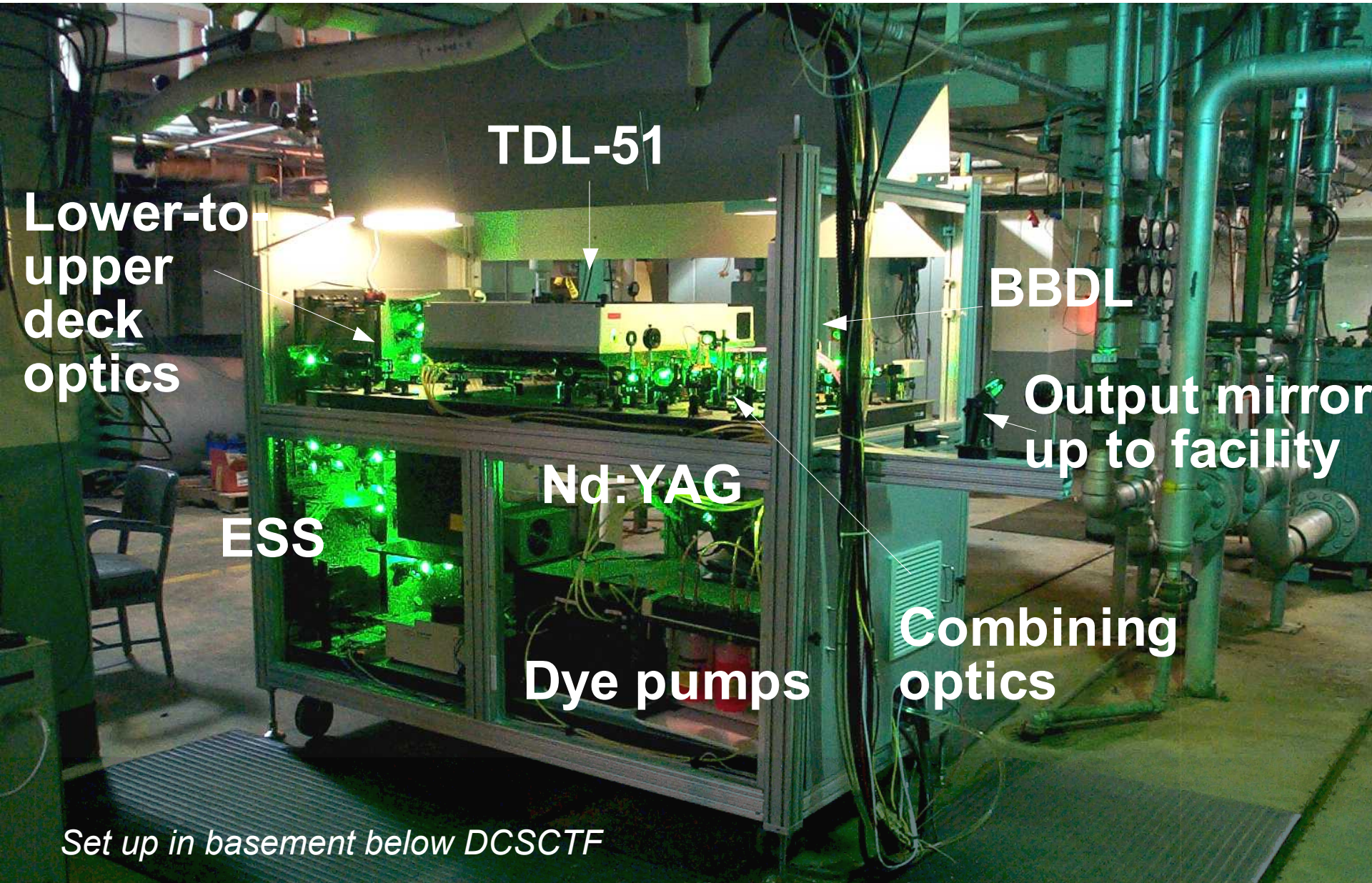




- Measurement obtained:
  - In UVa Scramjet (Conf C)
  - φ=0.18, Plane 2, z/H=0
  - in10 ns (instantaneous)
  - at 20 Hz repetition rate
  - Measurement volume = 1.7 x 0.2 x 0.2 mm^3

NASA/GWU Mobile CARS System

→ Simplifies/speeds set up in facility

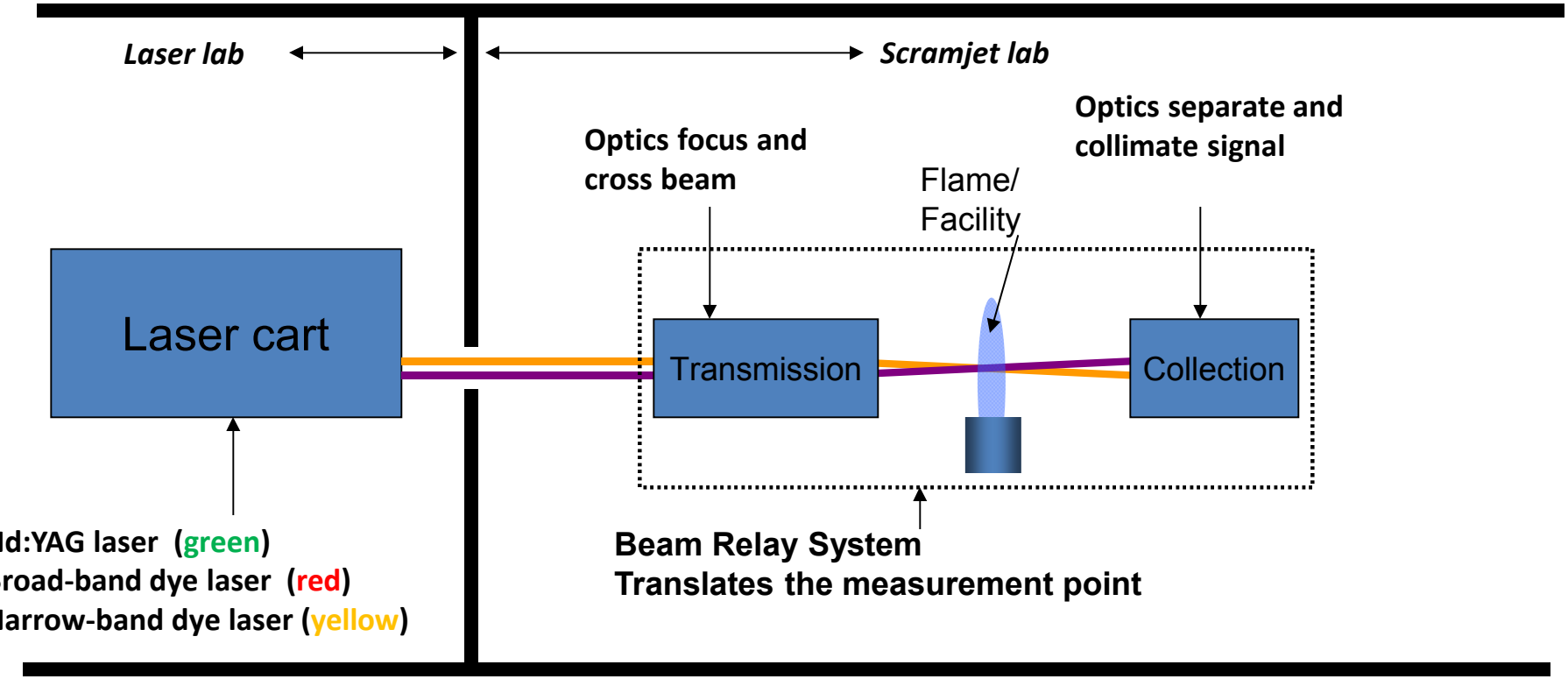


Set up in basement below DCSCTF

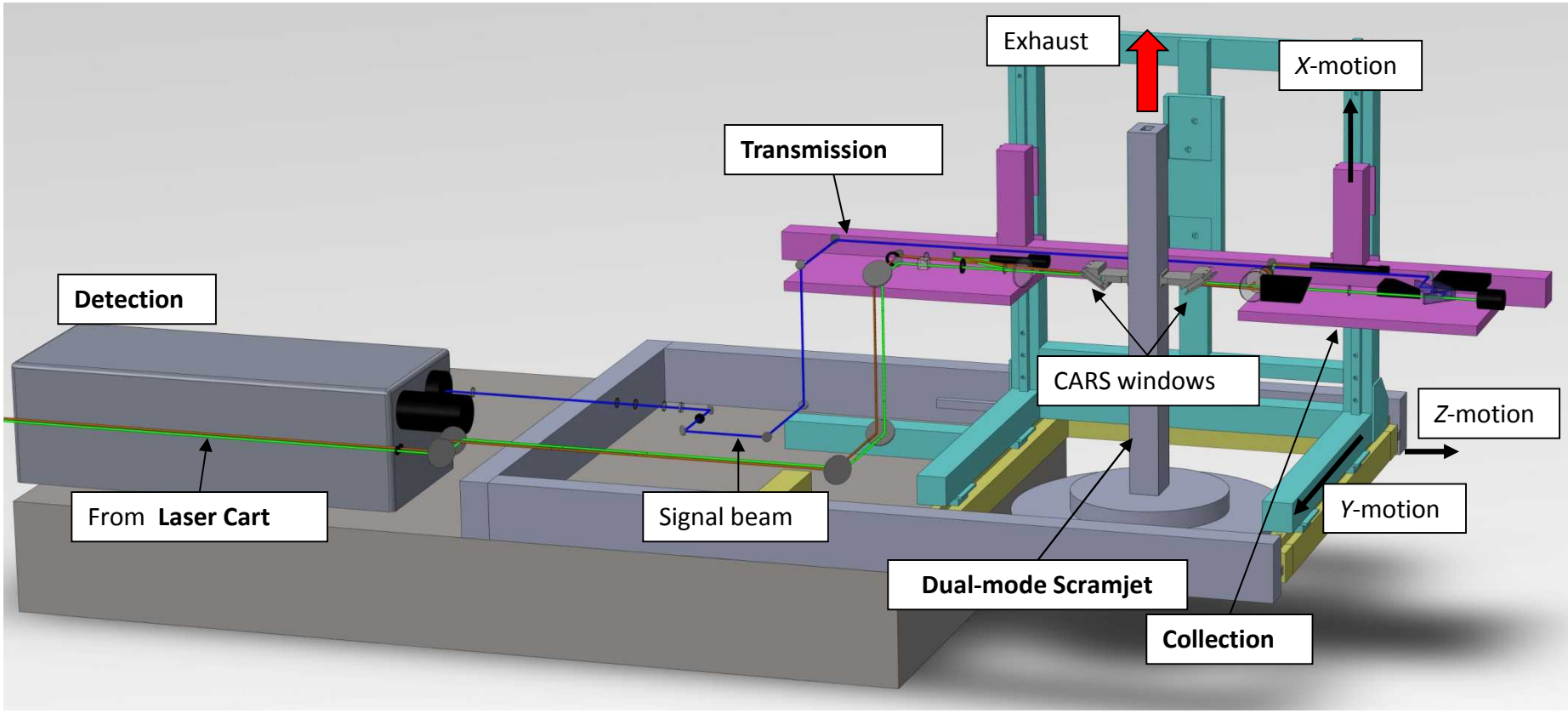
Features of NASA/GWU Mobile CARS System

- YAG laser plus 2 dye lasers on single cart
- Transmission and receiving optical stages
- Can put system on a truck and moved to facilities
- Measures T_{rot}, T_{vib}, N_2, O_2, H_2 mole fractions at 20 Hz.
  - Quantified accuracy, precision
- Fully automated data acquisition
  - Remote operation of system
  - X-Y (-Z) probe volume traverse capability
  - Input test matrix; then system acquires and automatically saves data for hours until complete
  - Laser power automatically adjusted to prevent saturation effects (detector saturation, Stark broadening, Raman pumping)
  - Beam alignment monitors; remote control alignment
- Beam shaping capability to maintain alignment measurement in turbulent/vibration flows
  - 10x improvement compared to USED CARS

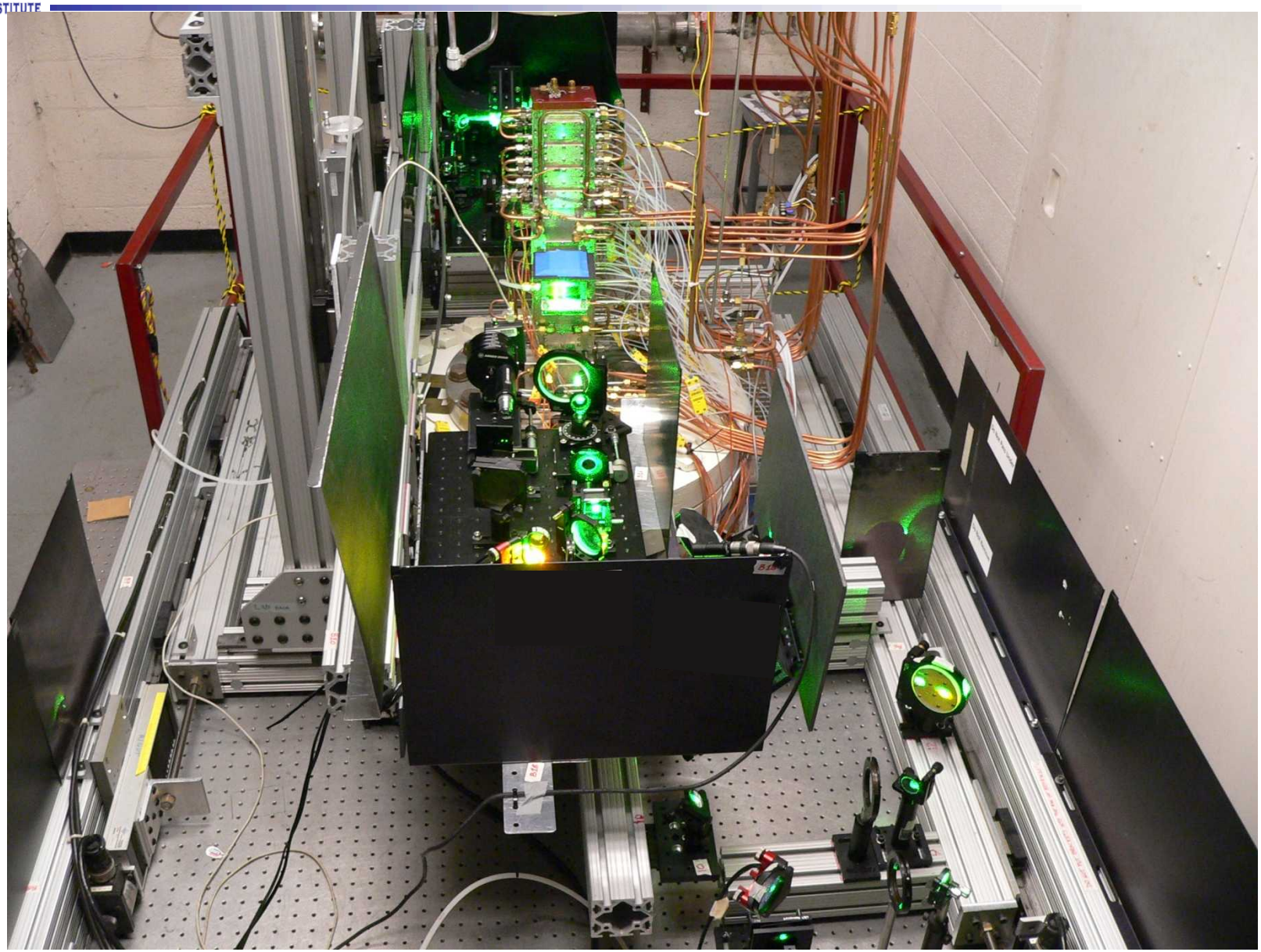
Setup at UVa: Optical System 1



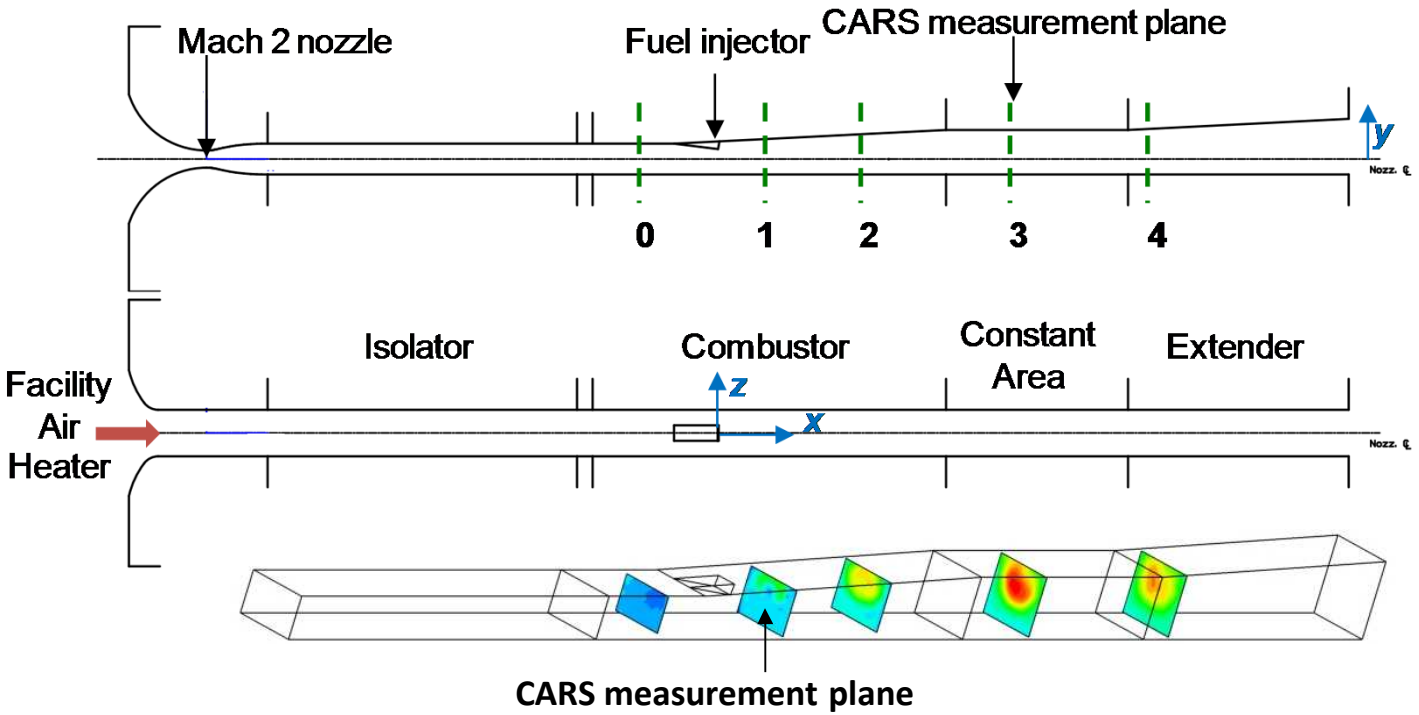
Setup at UVa: Optical System 2



Transmission stage and test section at UVa



Facility / Dual Mode Scramjet at UVa



- Electrically heated clean air, nominal T_t = 1200 K, M = 2 nozzle
- Long test times allow large data sets at steady inflow conditions
- Highly turbulent flowfield
- Good optical access

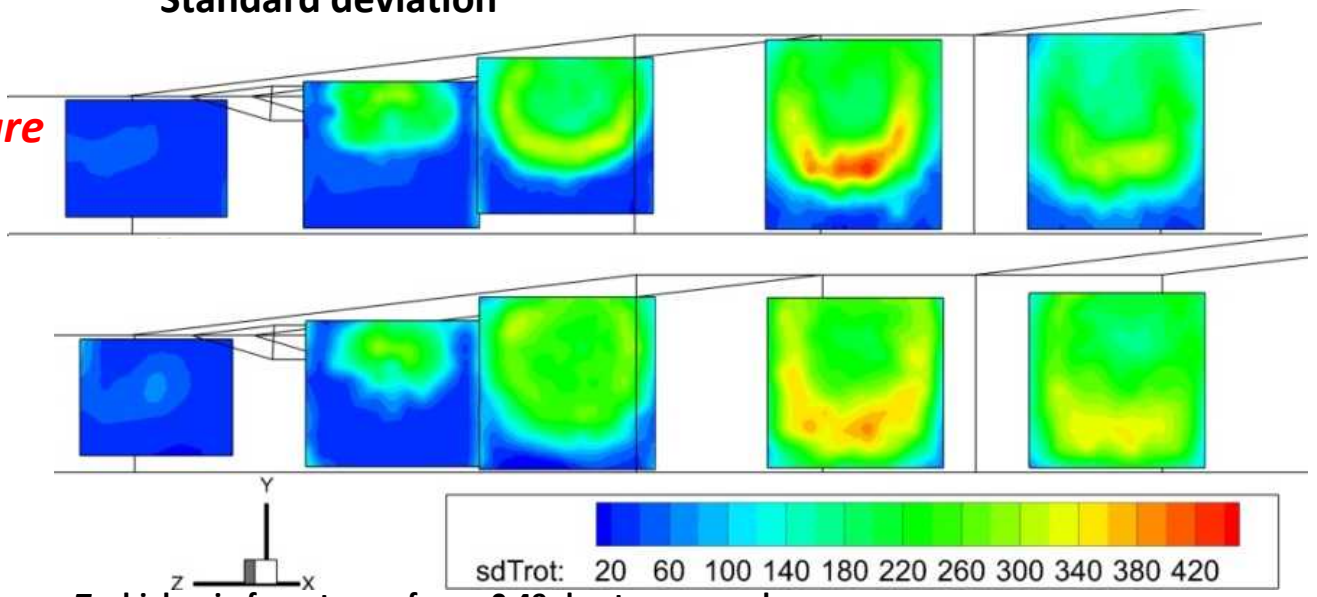
Mean and Standard Deviation Temperature

Standard deviation

Rotational Temperature

φ=0.18

φ=0.49

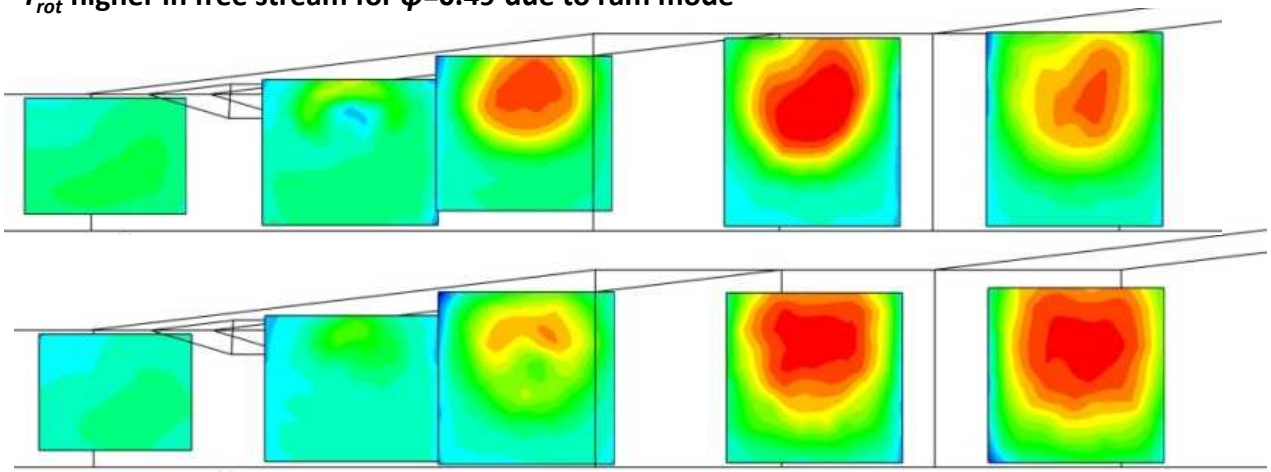


T_{rot} higher in free stream for φ=0.49 due to ram mode

Vibrational Temperature

φ=0.18

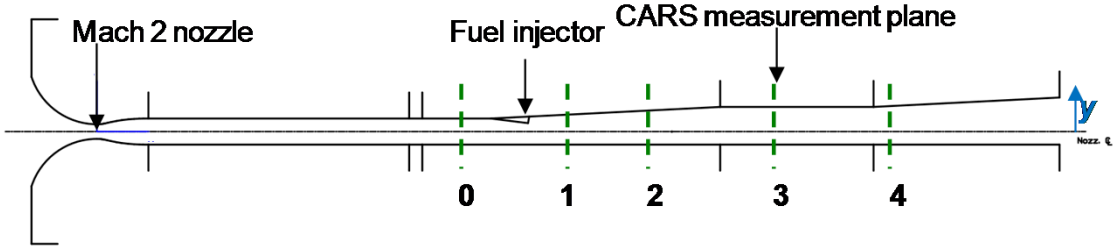
φ=0.49



- T_{N2vib} 600 K higher than T_{rot} in facility freestream (expected; agrees with computations)

Contours of Mole Fraction N_2

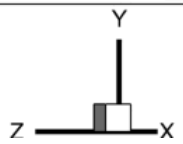
- Since inert shows overall mixing

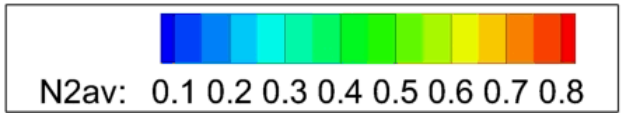


φ=0.18

φ=0.49

View from right looking upstream





CARS compared with CFD (LES from NC State)

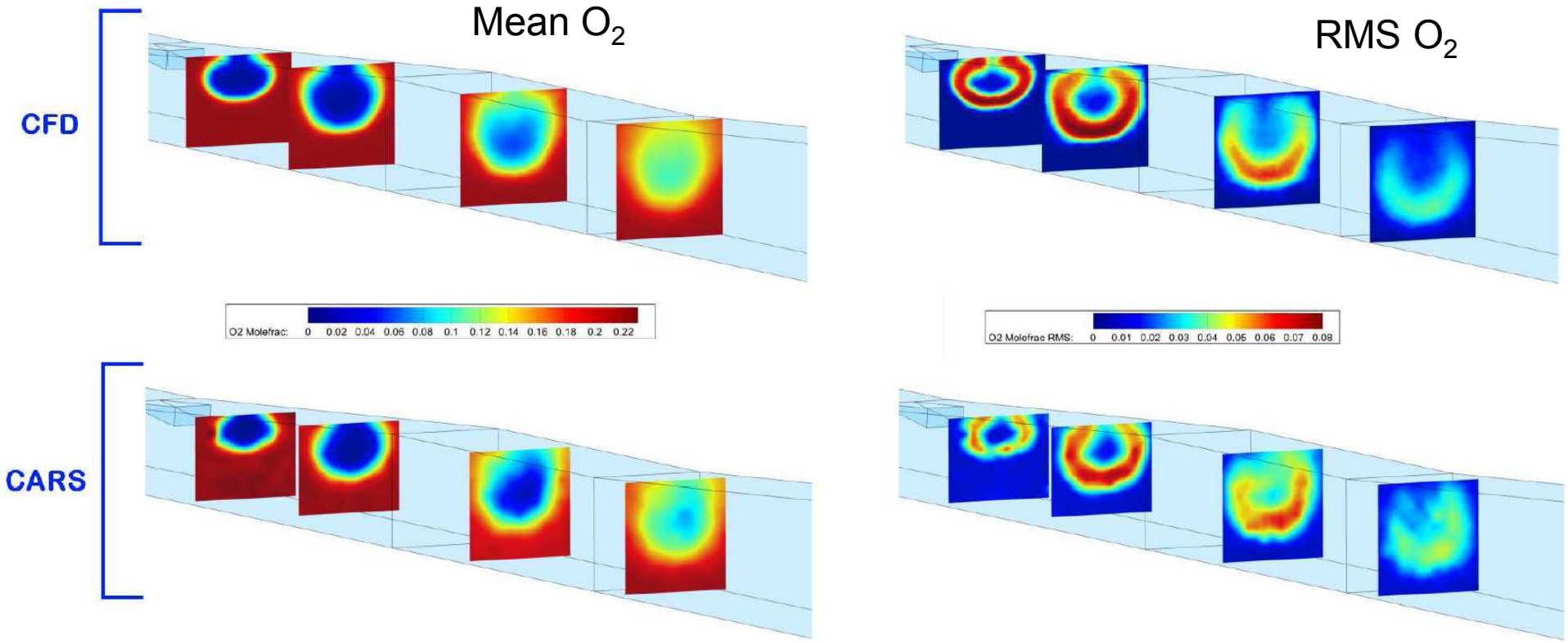
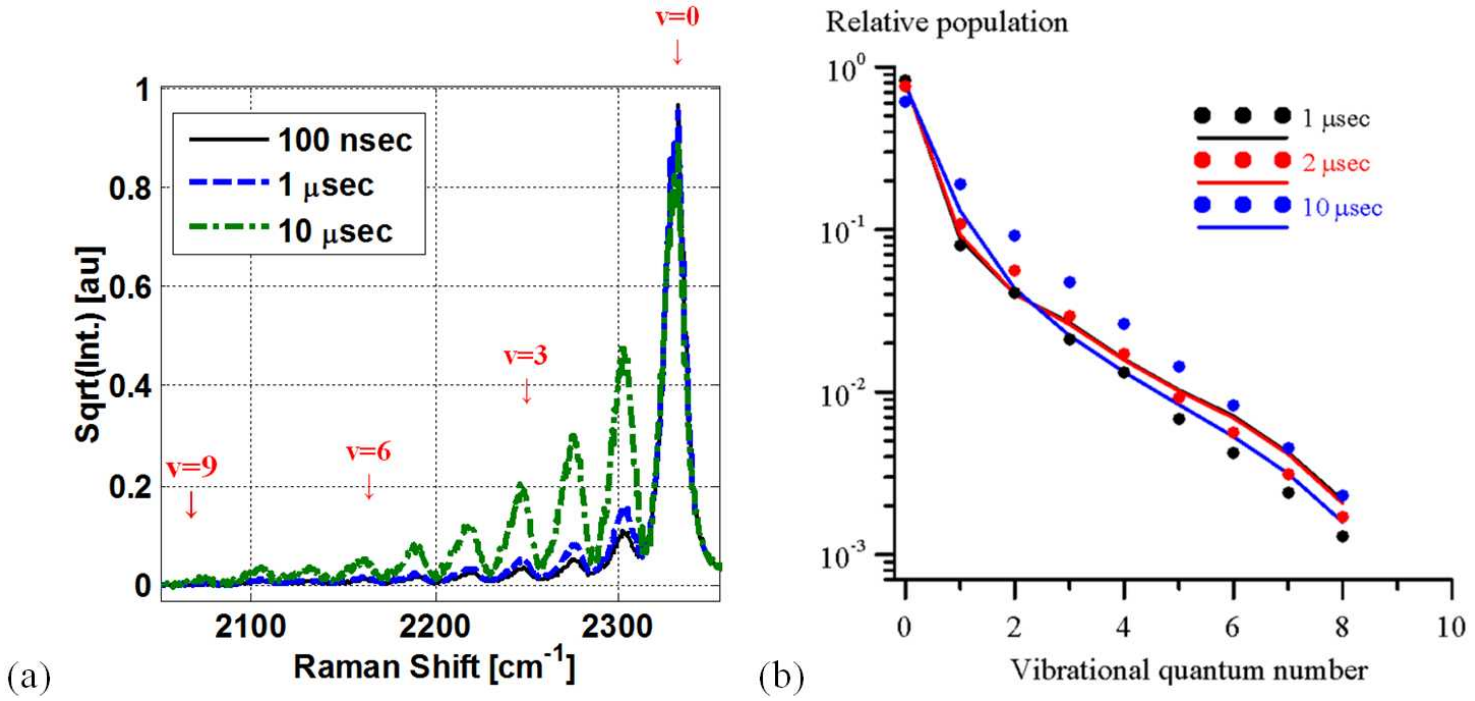


Figure 9. Comparison between CARS and O_2 concentrations and RMS fluctuations for '0.17' equivalence ratio.

- CARS provides valuable quantitative comparison data
  - Fulton, Edwards, et al, (Configuration C) AIAA-2013-0117, 2013

CARS for non-thermal plasma flow

- 100 Torr pulsed discharge at Ohio State



- CARS can measure individual state populations for model validation.

- A. Montello, Z. Yin, D. Burnette, I.V. Adamovich, and W.R Lempert, "Picosecond CARS Measurements of Nitrogen Vibrational Loading and Rotational/Translational Temperature in Nonequilibrium Discharges," J. Phys. D: Appl. Phys., Sept, 2013.

Additional Thoughts on CARS

- CARS is a relatively accurate and precise high temperature measurement technique in reacting flow
  - Accuracy 2-3%; Precision 2-3% (Magnotti, 2012)
- CARS does not require flow seeding
- CARS can be performed through small windows or slots but need access on 2 sides.
- It is a relatively complicated technique, theoretically and experimentally.
  - Takes months to analyze data carefully.
- Is being extended to much faster measurements ~10 kHz with fs CARS (Lucht and coworkers)
  - Very recently: 2 beam CARS, CARS imaging (Sandia)
- Has been performed using optical fibers to deliver the laser beams, both in ns and ps regimes

Conclusion

- Variety of different molecular-based measurement technologies available for measuring aerospace flows.
  - Can measure temperature, density, velocity, species concentrations, etc.
  - Which is best? Which technique to use?
- Must start measurement campaign with an interview of “customer” to determine the requirements
  - How do different measurement technique capabilities satisfy the requirements?

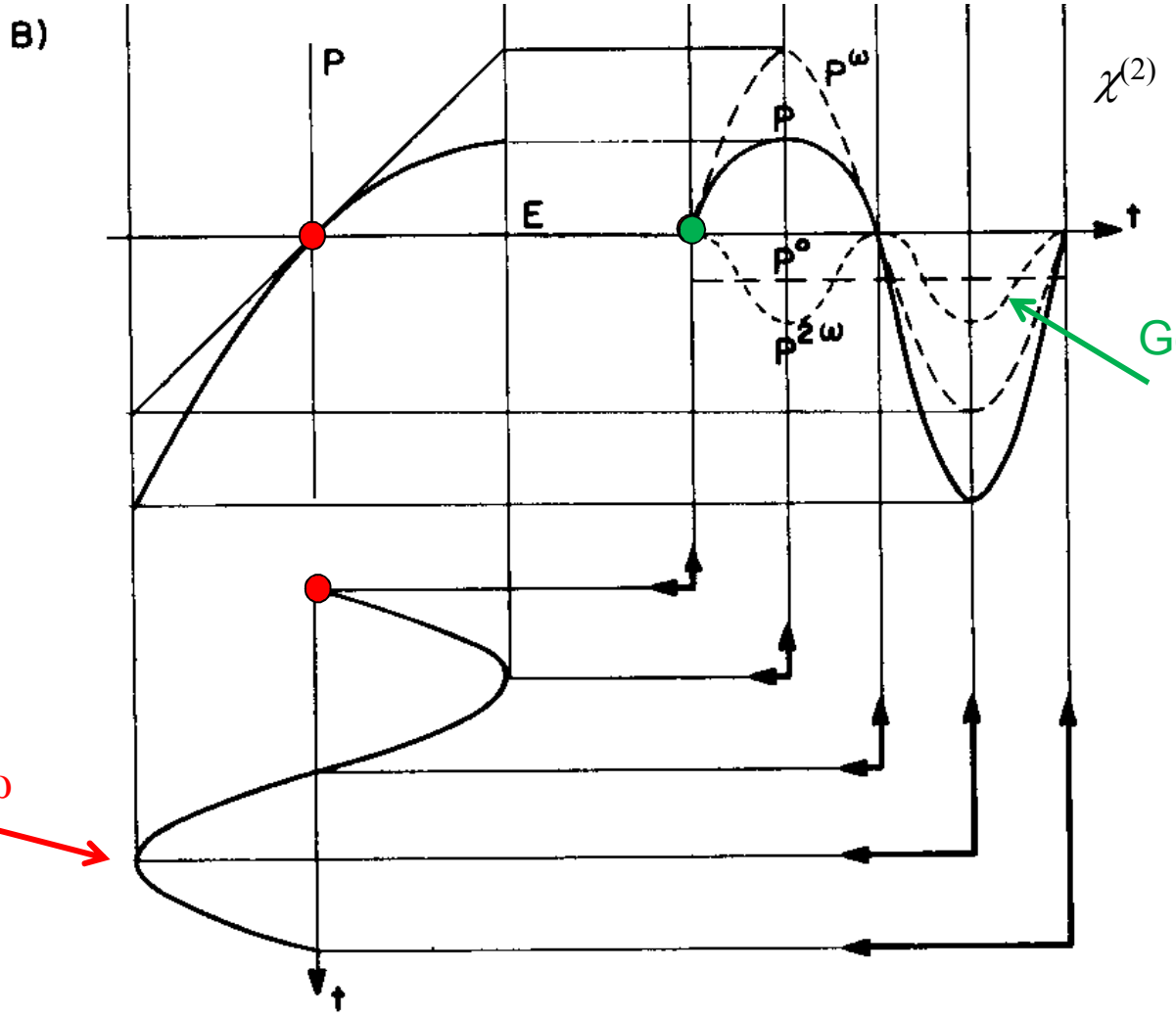
The interview prior to getting started

- What parameter(s) need to be measured?
- Must multiple parameters be obtained simultaneously to determine correlations?
- What spatial resolution is required?
- Is imaging required or are single-point or line measurements sufficient?
- What temporal resolution is required? (e.g. time required for a single measurement)
- Do measurements need to be time resolved? (e.g. a continuous sequence of data)
- What accuracy is needed?
- What precision is needed?
- What quantity of data is required?
- When is the data needed? Is instant (real-time) data required?
- Where in the flow are measurements required? (inflow, exit, near walls, etc.)
- What type of optical access is available?
- Can (toxic) seed gases be introduced? Will they influence the properties being measured?
- What is the ordered priority of the above requirements?

Conclusion

- Variety of different molecular-based measurement technologies available for measuring turbulent flows.
  - Can measure temperature, density, velocity, species concentrations, etc.
  - Which is best? Which technique to use?
- Must start measurement campaign with an interview of "customer" to determine the requirements
  - How do different measurement technique capabilities satisfy the requirements?
  - Also consider practical issues:
    - Experience of the research team
    - Available equipment / budget
    - Schedule, difficulty of method, etc.
  - Rarely a perfect match, but a match can provide the data needed to advance or validate theory or computations

Non-linear Susceptibility



G.C. Baldwin "An introduction to nonlinear optics." *New York: Plenum Press,* (1969).

- Non-linear effects: Intense electric fields of one frequency can generate a **P** at another