

Measurement techniques for combustion and high temperature flows

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Outline

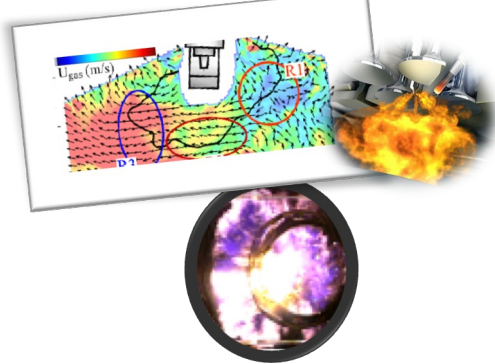
- Why and how we measure
- Fundamentals of optical diagnostics
 - Light sources and signal collection
 - Detection methods
 - Light-molecule interactions
- Incoherent
 - Scattering and particle velocimetry
 - Rayleigh scattering
 - Raman scattering
 - Absorption
 - Laser induced fluorescence
 - Laser-induced incandescence
- Coherent techniques
 - CARS
 - LIGS

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Turbulent combustion in real devices

IC engines



unsteady
autoignition
high
pressure
sprays
soot, NO_x,
CO

Gas turbines, rockets



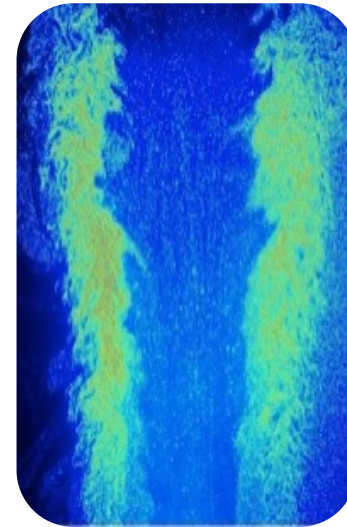
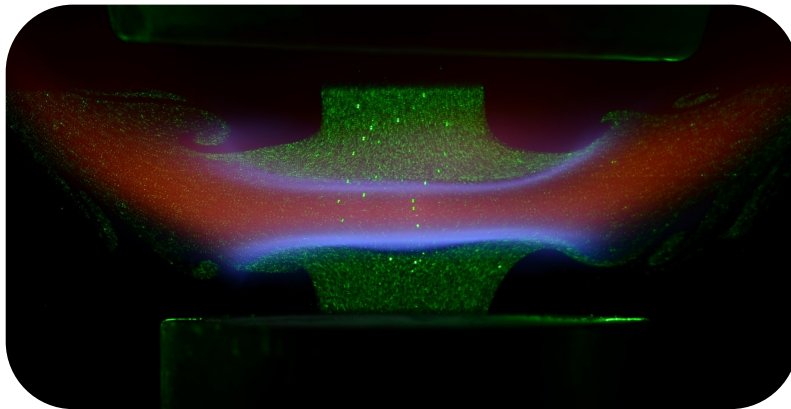
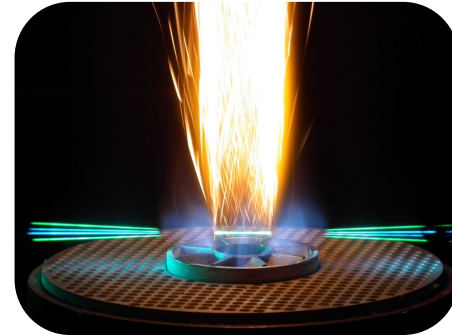
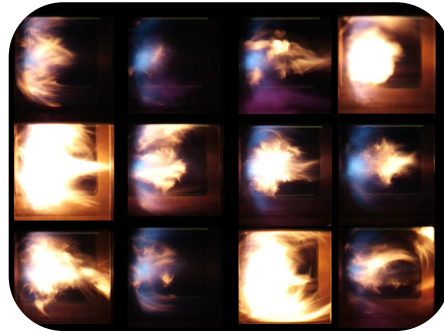
blow off
instabilities
high pressure
sprays
soot, NO_x, CO

Furnaces and industrial processes



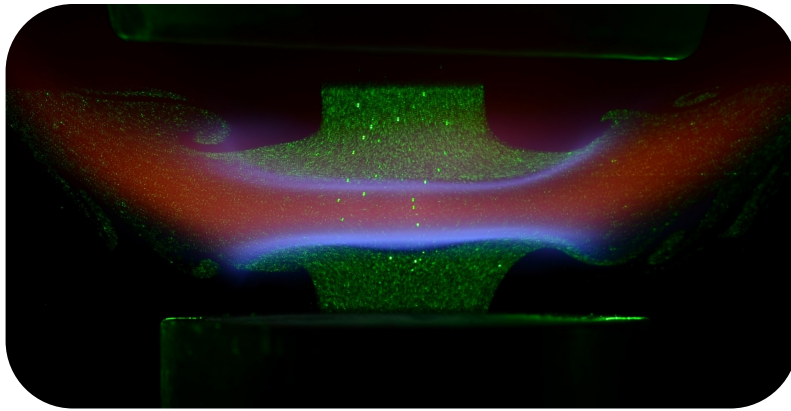
solid particles
radiation
soot, NO_x, CO

Bringing reacting flows to the lab

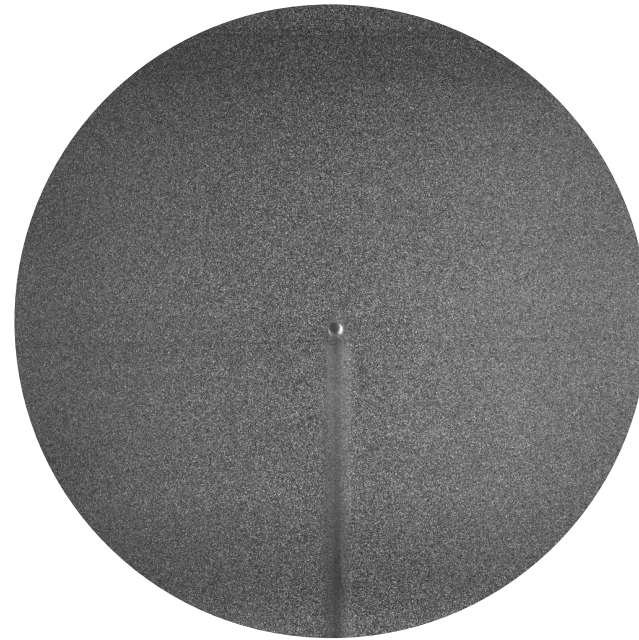


Many spatial and time scales, many scalars

Reacting flows: laminar

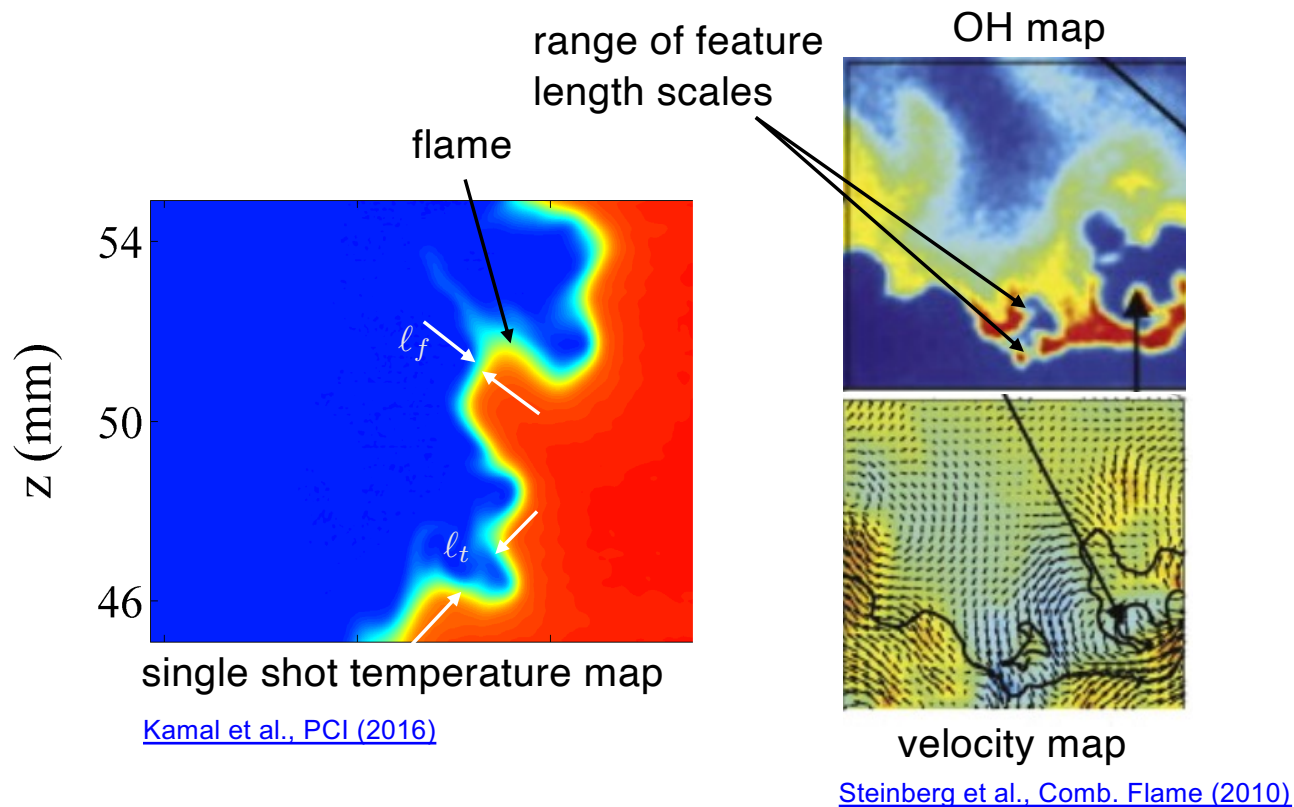


D. McGrath, U. Cambridge



Courtesy: B. Renou,
INSA-Rouen

Resolving turbulent and flame scales



Spatial scales from μm to tens of cm
Time scales from μs to s

$$\begin{matrix} \ell_t & \tau_t & u_t \\ \ell_f & \tau_c & S_L \end{matrix}$$

Big whirls have little whirls,
That feed on their velocity;
And little whirls have lesser
whirls,
And so on to viscosity
Lewis F. Richardson

$$\varepsilon = \frac{u_t^2}{\tau_t} = \frac{u_t^3}{\ell_t} = \frac{u_\eta^3}{\eta} = \frac{\nu}{\tau_\eta^2}$$

large $\rightarrow$ small

Why we measure

models

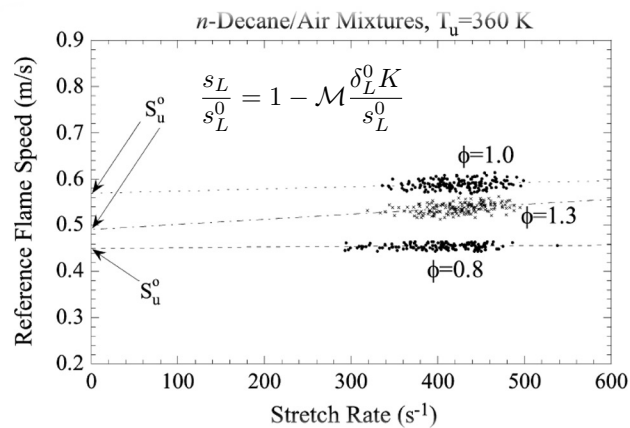
$$\rho \frac{\partial \varphi_i}{\partial t} + \rho \mathbf{v} \cdot \nabla \varphi_i = \nabla \cdot (\rho D_i \nabla \varphi_i) + w_i$$

$$\frac{\partial \rho \mathbf{v}}{\partial t} + \nabla \cdot (\rho \mathbf{v} \mathbf{v}) = -\nabla p + \nabla \cdot \boldsymbol{\tau} + \rho \mathbf{g}$$

solution

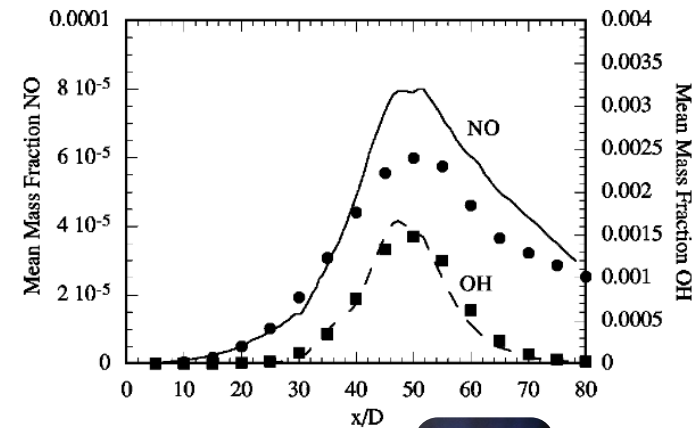
$$\phi_i, \mathbf{v}$$

Laminar flame



$$\phi_i, \mathbf{v}$$

validation



Sandia flame D

Kumar, K., Sung, C.-J.
Comb. Flame 151: 209
(2007)



H. Pitsch and H. Steiner
Physics of Fluids,
12 2541 (2000)



Brazil Winter School 2025

Things we would like to measure

Flow field

- Mean, fluctuations, correlations of velocity (1-2-3 components)
- Strain, vorticity (gradients in 2D or 3D)
- Pressure and fluctuations
- Length and time scales of turbulence, coherent structures

Scalar field

- Mean, fluctuation of temperature, major and minor species concentrations
- Equivalence ratio or mixture fraction
- Scalar gradients and length scales
- Structural information based on 2D- or quasi 3D-diagnostics
- Heat release rate (where, when, intensity)

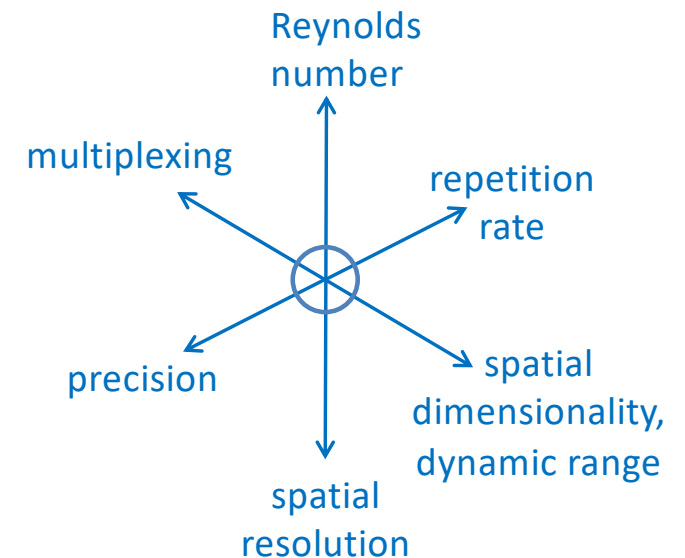
Multiphase flow and particulates (e.g., coal, spray, soot, synthesis)

- Particle size, number density, volume fraction, velocity, ...

Boundary and inflow conditions

- Flow and scalar fields
- Surface temperatures

Everything everywhere all at once...



\$\$\$

Choosing the measurement technique

Time scale

long

short

Spatial resolution

coarse

fine

Accuracy

average

high

Selectivity

low

high

Cost

low

high

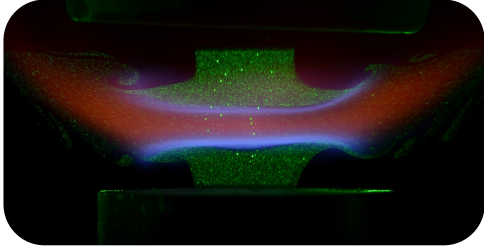
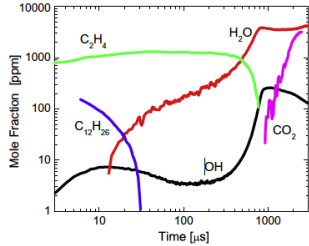
Interference

intrusive

non-intrusive

Right tool for the right job
Focus on: what is the question?

Examples

	ΔL	Δt
	Resolving temperatures in a steady laminar flame	$\sim 10\text{-}100\ \mu\text{m}$ $\sim \text{s}$
 Fig. 3. Species time-history measurements for <i>n</i>-dodecane oxidation. Nominal initial conditions: 1410 K, 2.3 atm, 457 ppm <i>n</i>-dodecane/O₂/argon, $\phi = 1$. Davidson et al., PCI33, 2011	Measuring species during a shock tube reaction	$\sim 1\ \text{cm}$ $\sim 10\ \mu\text{s}$
	Instantaneous temperatures in a turbulent flame	$< 50\ \mu\text{m}$ $< 1\ \mu\text{s}$

Intrusive probe measurements



Hotwire

10^{-3} s

(<500-800 Hz)

10^{-3} m



Thermocouples

10^{-2} s

(100 Hz)

10^{-3} m



Sampling probe

10^1 s

(0.1 Hz)

10^{-2} m



Pressure sensor

10^{-4} s

(<15 kHz)

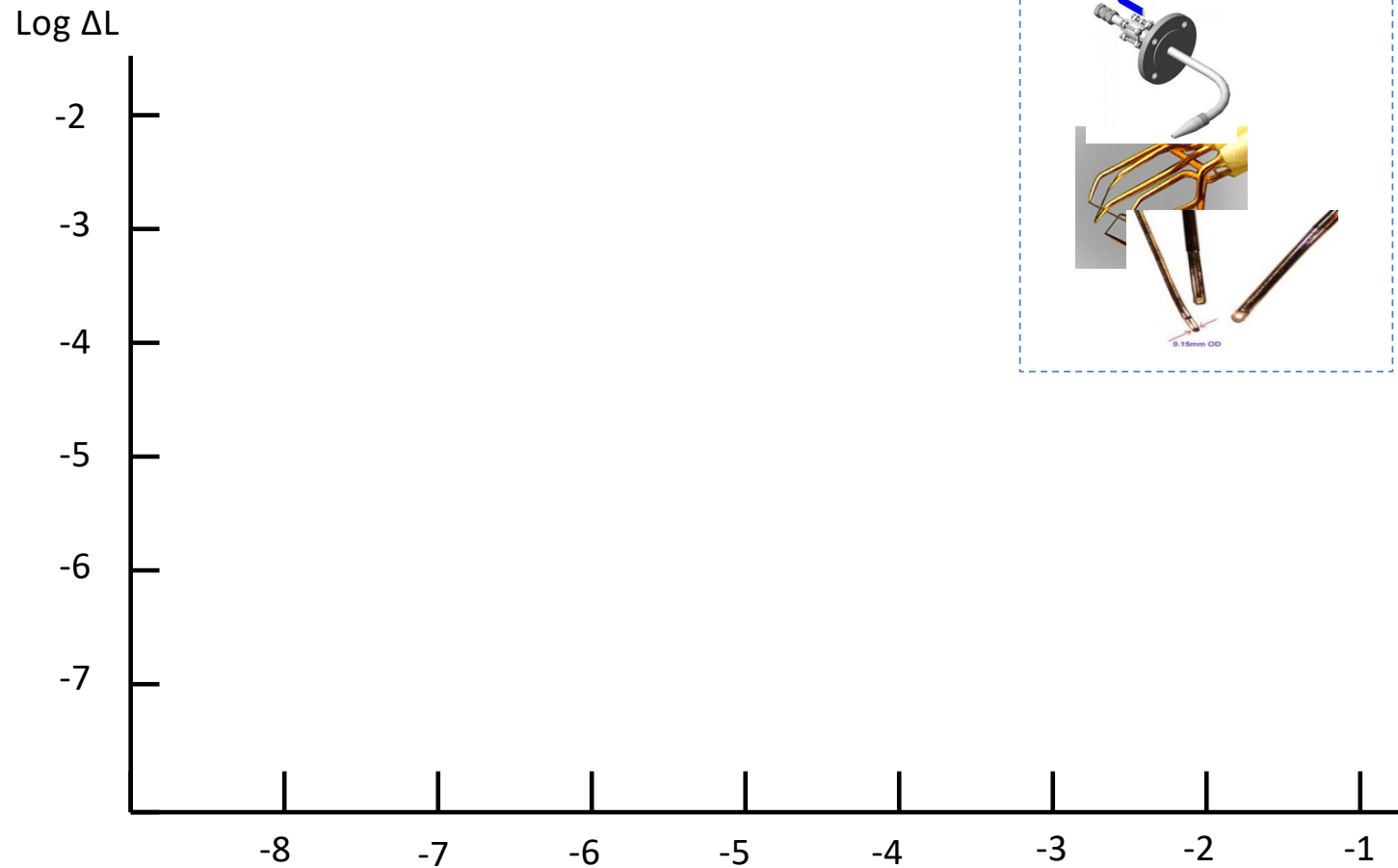
10^{-2} m

- Often **affect or are affected by sampling region:** accuracy
- **Limited time response** and resolution due to size and time lags in lines
- **Material limitations** : cannot cope with high temperature environments

Optical measurements

- Require optical access
- Significant effort in setup and instrumentation

Intrusive or optical methods?



Time and spatial resolution

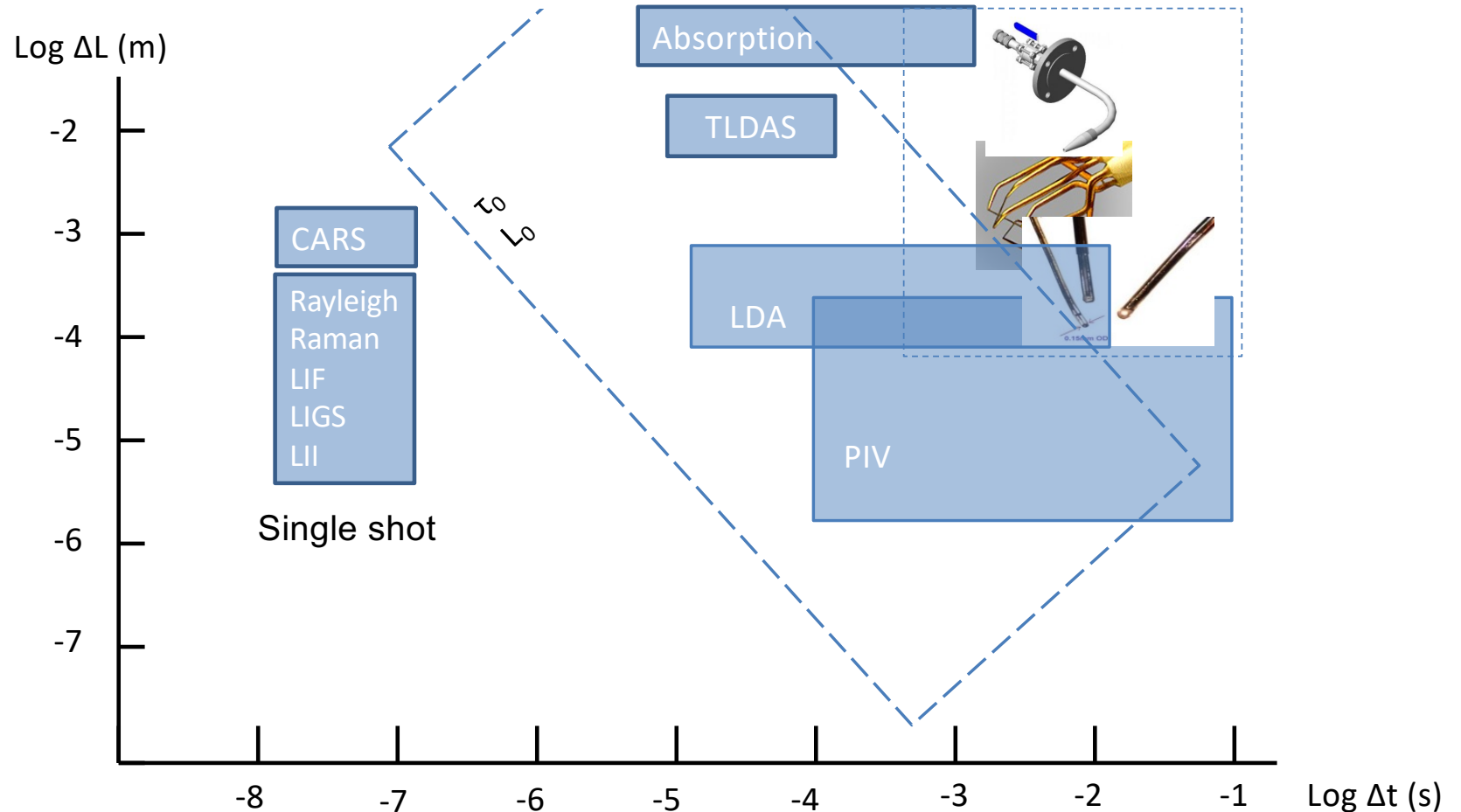
Optical methods :

- Non-intrusive
- Fast
- High resolution

Optical methods :

- Require access
- Expensive
- Expertise

Always use
simplest method
compatible with
the question to be
answered!



Questions



What is the question that you are trying to resolve?



What is the hypothesis?



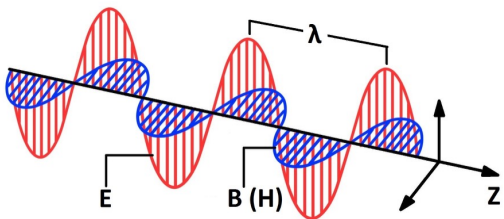
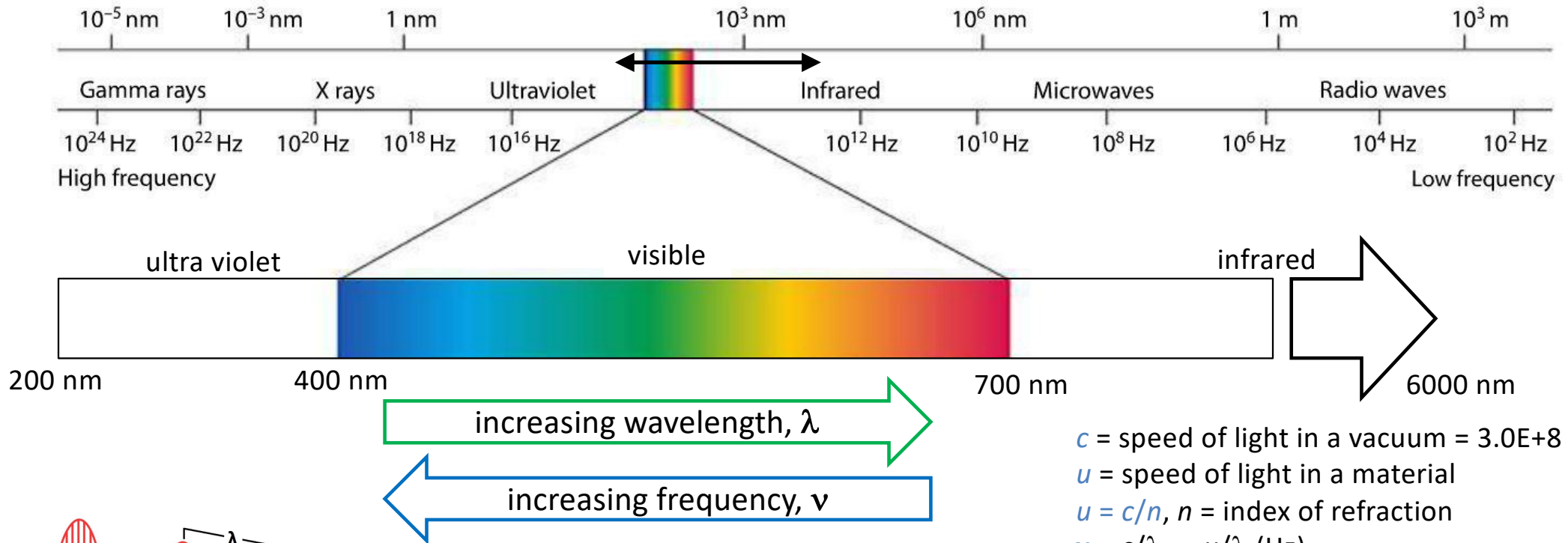
Do you have a model to compare results?

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Electromagnetic radiation

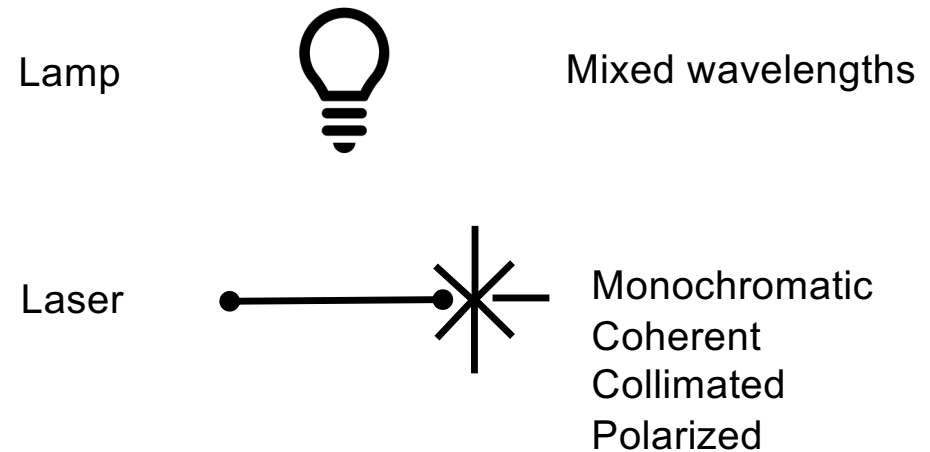
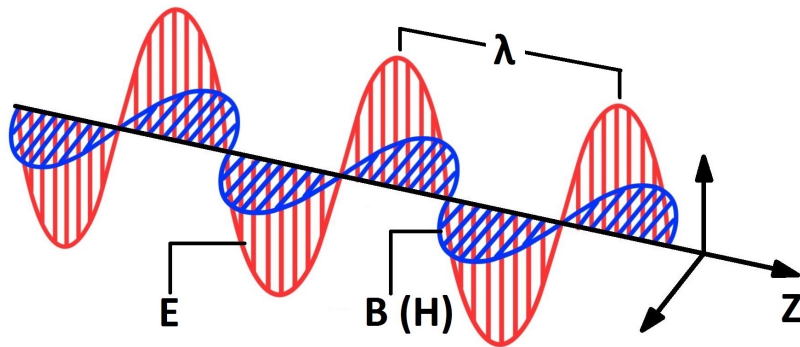
Most combustion diagnostics: $200 \text{ nm} < \lambda < 6000 \text{ nm} = 6 \text{ }\mu\text{m}$



c = speed of light in a vacuum = $3.0\text{E}+8 \text{ m/s}$
 u = speed of light in a material
 $u = c/n$, n = index of refraction
 $\nu = c/\lambda_0 = u/\lambda \text{ (Hz)}$
 $E = h\nu$ = photon energy (J)
 $h = 6.63\text{E}-34 \text{ J}\cdot\text{s}$ (Planck's constant)

Note: Spectroscopists often use wavenumbers (cm^{-1}) = ν/c or = $10,000,000/\lambda_0$ with λ_0 in nm

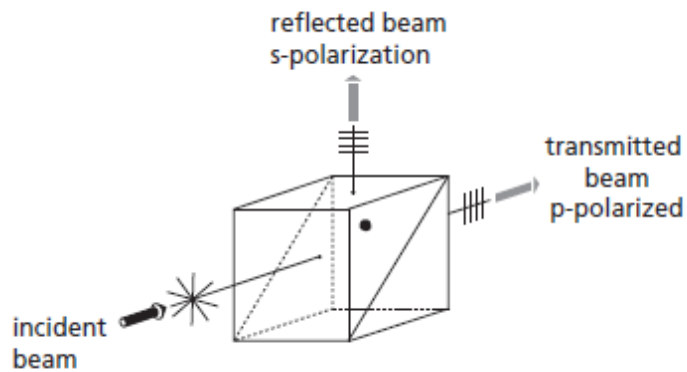
Light sources, lasers and all that



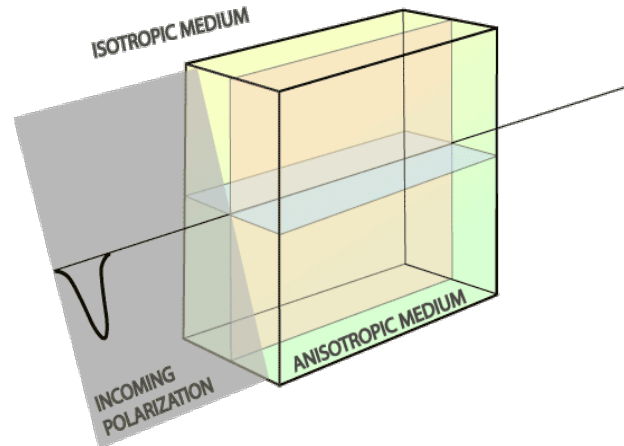
Beam propagation:

- **Monochromatic**: Single wavelength
- **Polarized**: linear/circular/random wave plane orientation
- **Coherent**: all waves in phase
- **Collimated**: all waves propagate in parallel

Waves can be manipulated using polarization



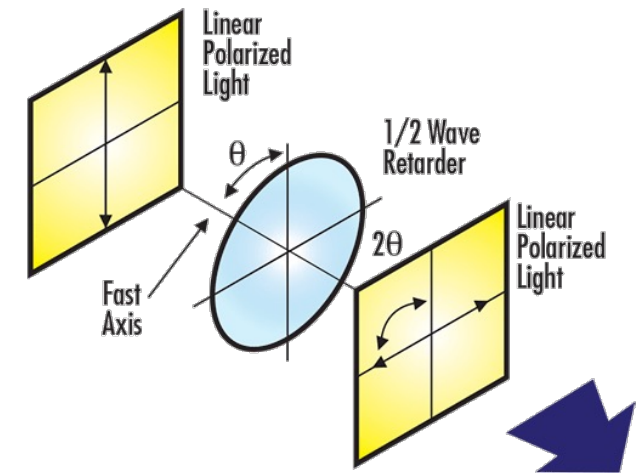
A **polarizer** separates an incident beam into two linear components



<https://en.wikipedia.org/wiki/Birefringence>

A **birefringent material** (e.g., crystal) has a different index of refraction parallel vs. perpendicular to the optical axis of the material. Components travel at different speeds

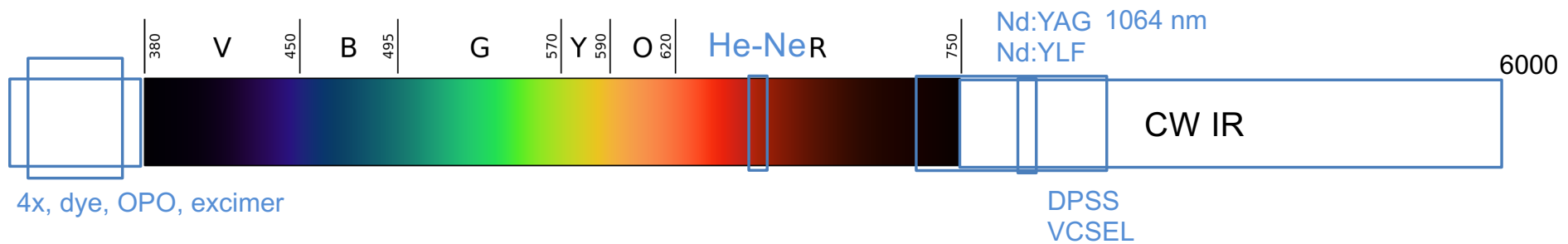
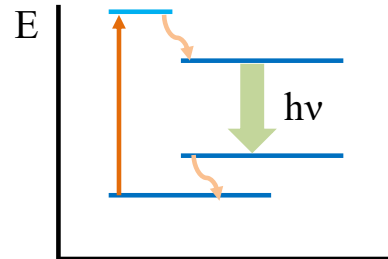
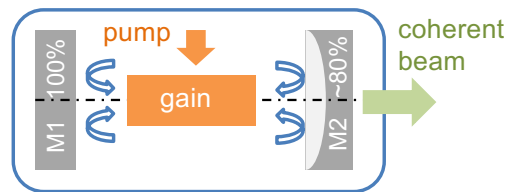
<https://www.edmundoptics.com/resources/application-notes/optics/understanding-waveplates/>



Example: a **half wave plate** rotates the polarization of a linearly polarized beam by twice the angle between its optical axis and the polarization axis of the incoming beam

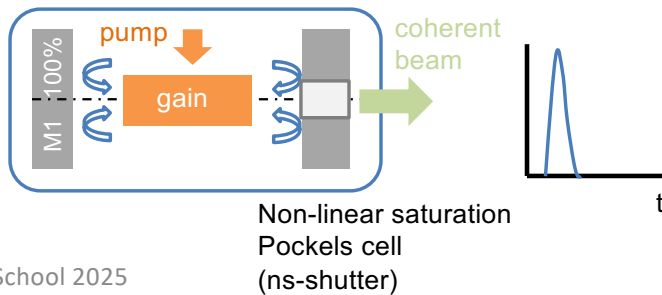
Combine a half wave plate with a polarizer to make a **variable beam splitter** or **variable attenuator** for a linearly polarized laser beam.

Light Amplification by Stimulated Emission of Radiation



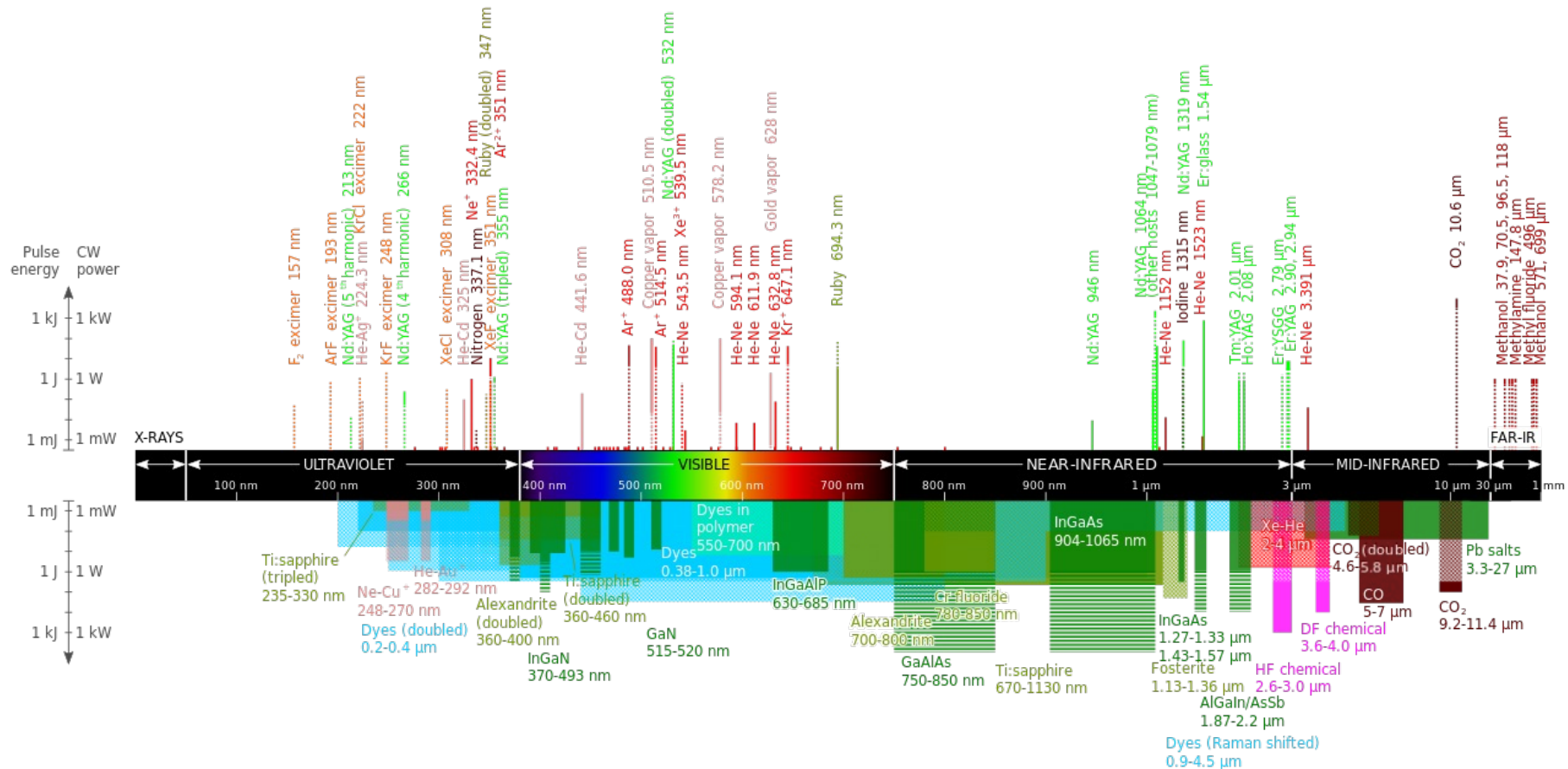
Covered by harmonics, OPO and frequency mixing

Pulsed



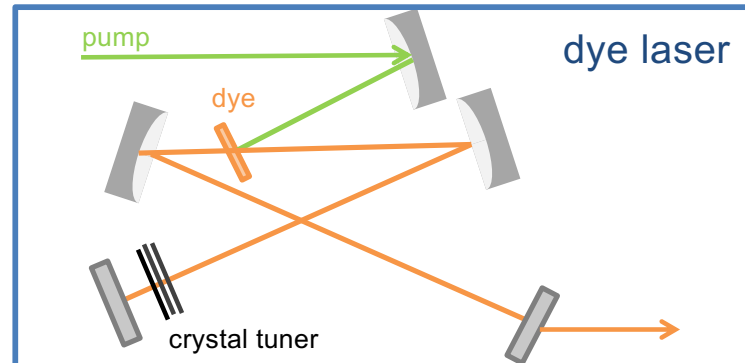
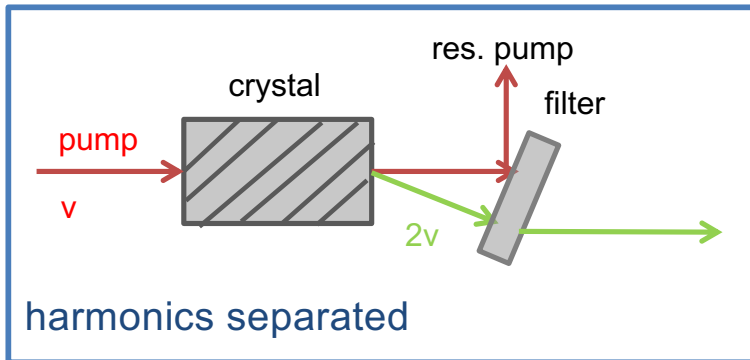
Property		
Operation	Continuous	Pulsed
Power	W	kW-GW
Time interval	μs	ns, fs, ps
Wavelength	Vis-IR	Visible-near IR

Different types of lasers for different jobs

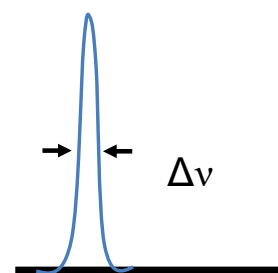
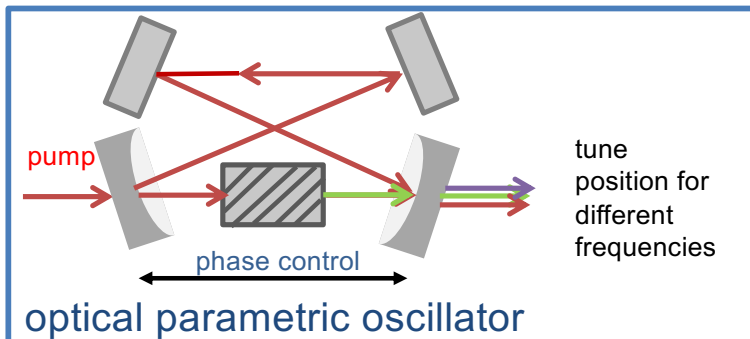


[Weber, Marvin J. Handbook of laser wavelengths, CRC Press, 1999](#)
Adapted by Danh.

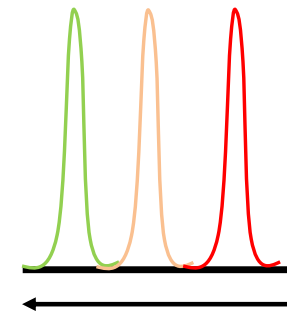
Reaching different wavelengths



UV to near IR
By changing dyes and/or
adding beam combiner



Line width:
OPO : $<1 \text{ cm}^{-1}$
Dye: $<0.01 \text{ cm}^{-1}$



Tunability

Conversion
efficiency

Recent developments:
fs/ps lasers pulses: wide bandwidth:

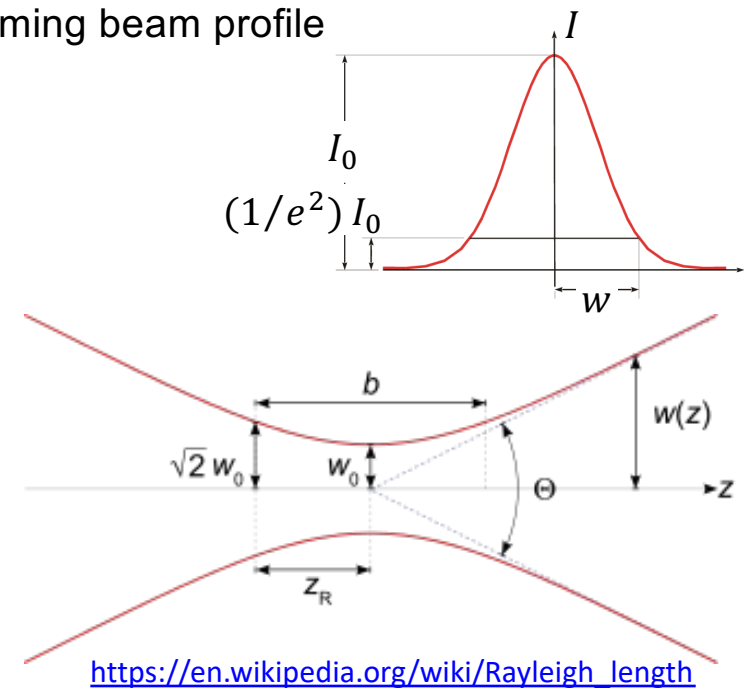
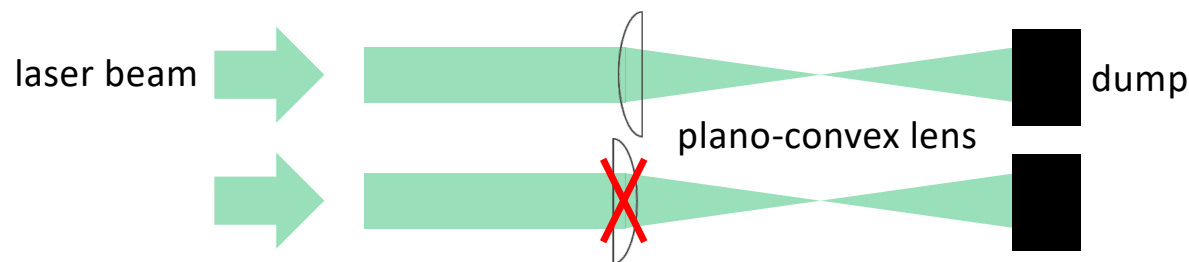
$$\Delta t \Delta e < \frac{h}{2\pi}$$

Energy conservation
phase matching

$$\nu_1 + \nu_2 = \nu_3$$

Optics (one-pager)

- What's at the focus?
 - Profile at the focus is the **spatial Fourier transform** of the incoming beam profile
 - Gaussian beam (best possible profile, TEM_{00})
 - **Gaussian beam waist**, $D = 2w_0 \cong 4\lambda/\pi\theta$
 - Rayleigh range, $z_R = \pi w_0^2/\lambda$
 - Estimate laser sheet width at center and edge on image
 - Typical laser beam does not focus as well
 - Incoming top hat beam is a sinc function (rings) at the focus
 - Typical focusing range in the visible range is $\sim 100 \mu\text{m}$
- Right way and wrong way (let both surfaces do the work)



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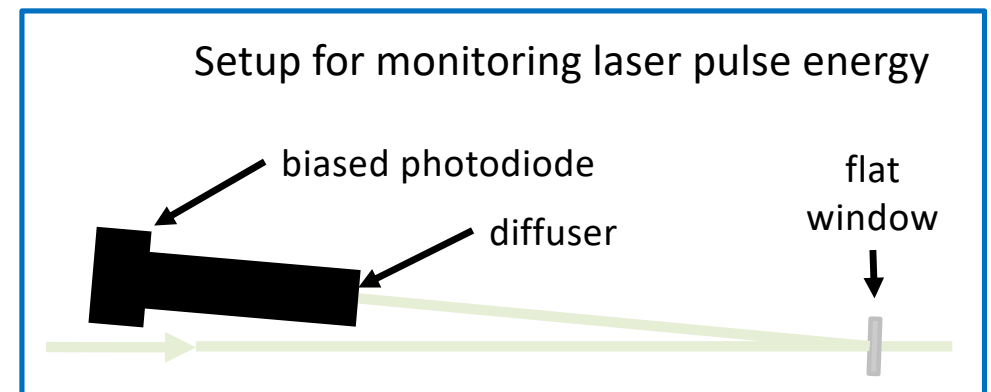
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Photodiodes

- Semiconductor devices that convert light to current
- Fast ($\sim$ ps)
- Diagnostic applications:
 - Laser absorption measurements
 - Laser power/energy monitoring
 - CW and pulsed
 - Chemiluminescence
 - ...



Material	Wavelength Range (nm)
Silicon	190-1100
Germanium	400-1700
Indium gallium arsenide	800-2600
Lead sulfide	1000-3500
Mercury cadmium telluride	400-14000



Photomultiplier tubes

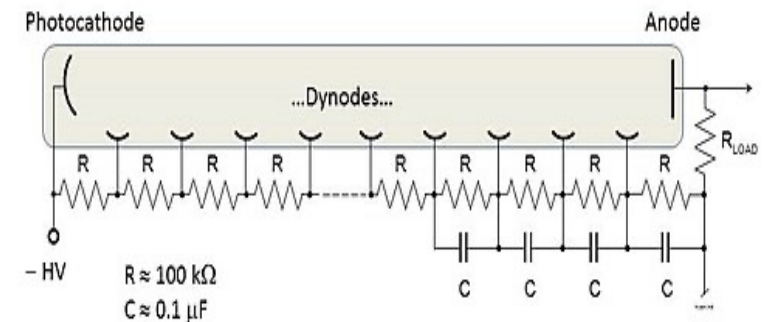
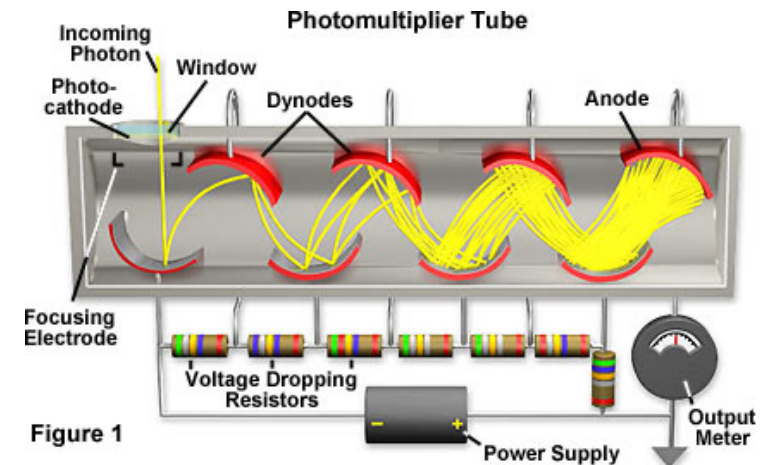
- Good for low level signals (even photon counting)
- Electron cloud is amplified through a dynode chain (~10 stages)
- Custom socket design to optimize performance for pulsed measurements: sensitivity, dynamic range, linearity
- Different photocathode materials for different wavelength ranges
- Can be used as **fast shutters (ns range)** by modulating dynode voltage
- Combustion diagnostic applications:
 - **Pointwise** LIF and Raman/Rayleigh
 - LDA
 - LII
 - LIGS

https://en.wikipedia.org/wiki/Photomultiplier_tube

side-on

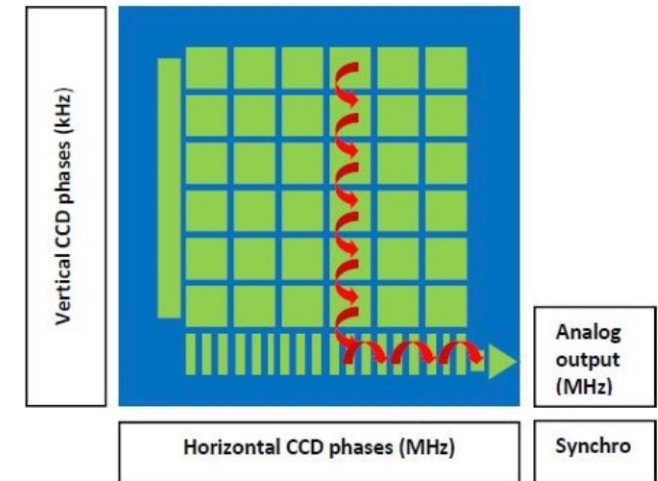


end-on



Array detectors (cameras): Fast moving technical area

- Main types in combustion diagnostics: CCD, EMCCD, CMOS, sCMOS by the way in which charge is generated/transferred
- **CCD = charge coupled device**
 - Detector of choice for most low rep-rate (CW or 10 Hz) imaging and spectroscopy applications
 - Pixels are read out sequentially through the same circuitry (*slower, more accurate*)
 - Back-illuminated architecture for high (~90-95%) quantum efficiency
 - Thermoelectric cooling to -100 C (cryogenic to -110 C) *minimizes dark current* (determines noise floor)
 - Programmable on-chip binning to reduce readout noise with low-level signals (example: spatial and spectral integration in Raman)
- **EMCCD = electron multiplying CCD**
 - Allows charge on each pixel to be multiplied before readout; helps with low signal levels, “*single photon sensitivity*”
- **CMOS = complementary metal oxide semiconductor**
 - Each pixel has its own amplifier, readout circuitry (*faster*)
 - High-speed imaging applications (PIV or PLIF at 10+ kHz)
- **sCMOS = scientific CMOS**
 - Significant improvements in QE, linearity, faster framing than CCDs



Minimum shutter ~ 1-10 μ s

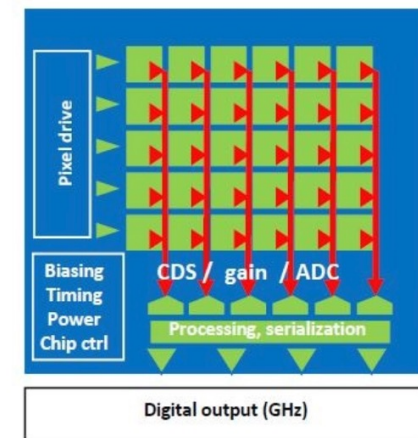


Image intensifiers

- Placed in front of CCD or CMOS detectors
 - **amplify** low light signals
 - allows **very short gate times (5-10 ns)** to **reject luminosity** from flame (shorter costs more)
- Main elements
 - Photocathode – converts photons to photoelectrons
 - Micro channel plate (MCP) – amplifies electron charge through each micro channel
 - Phosphor – converts electrons back to light at a good wavelength for the detector array
- Lens coupled or fiber-optic tapered bundle
 - Lens coupling gives better image quality, allows easy replacement of intensifier, bigger footprint
 - Fiber bundle gives good optical efficiency and compact design, but image quality is not as good (fixed honeycomb pattern, lower resolution)
- Increased gain helps signal-to-noise ratio up to a point, but produces **speckle noise** and **saturates easily**: not a general fix for everything

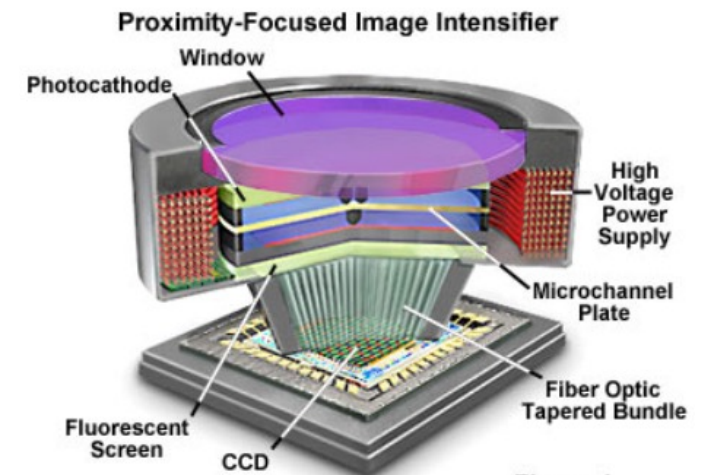
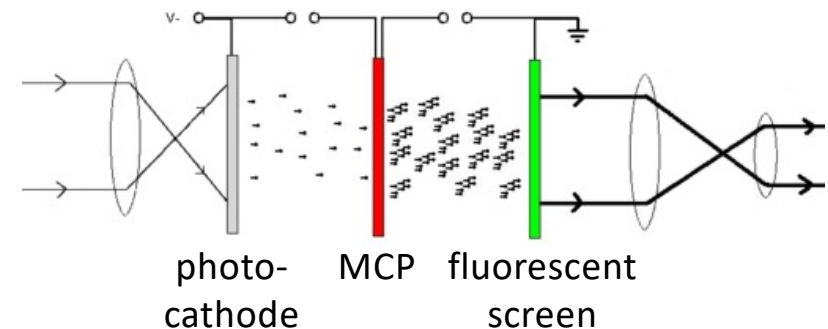


Figure 1

<http://hamamatsu.magnet.fsu.edu/articles/proximity.html>



https://en.wikipedia.org/wiki/Image_intensifier

Questions



What are the characteristic time/spatial scales of my problem?



Should I use an optical technique?



What quantities am I trying to measure?



Are they measurable?



What kind of wavelengths would be involved?



Do I need to use a laser?



Can I use natural emission?

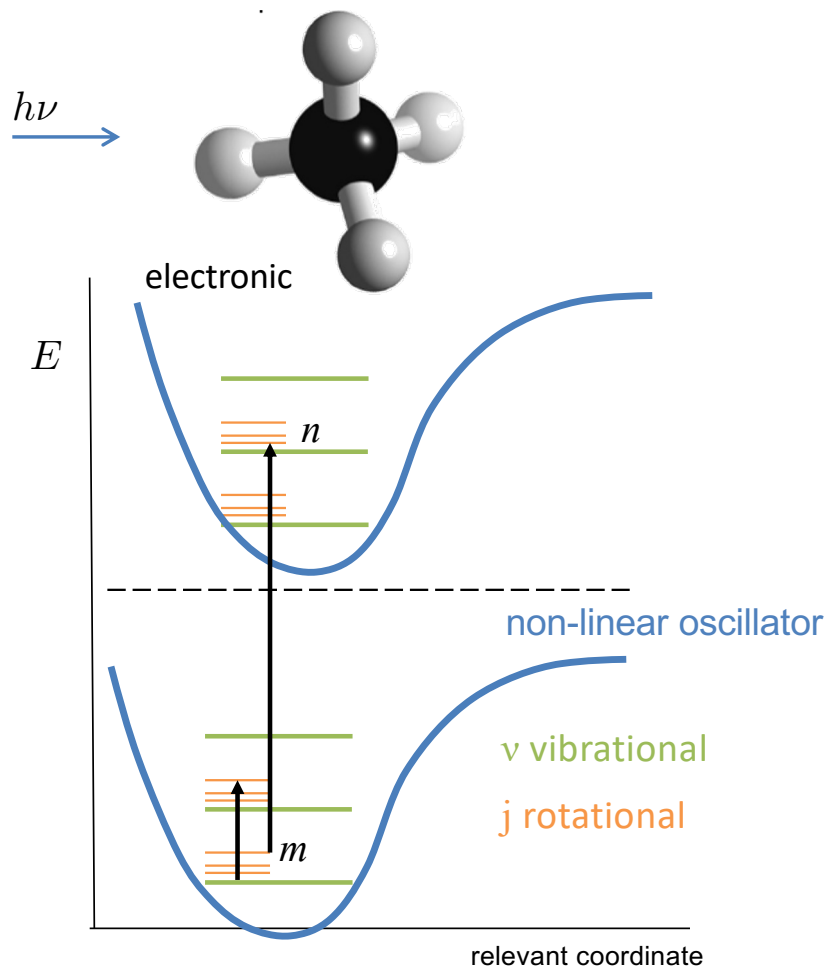


What other questions should I ask?

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Principles of radiation-molecule interactions



Atoms: spin, electronic

Molecules: spin, rotational, vibrational, electronic

$$\epsilon_{nm} = h\nu \quad h = 6.63 \times 10^{-34} \text{ J.s}$$

Molecules absorb or release photons

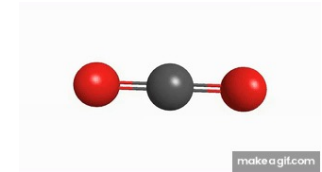
Energy levels are quantized

Levels determined as eigenvalues of the oscillation for each mode

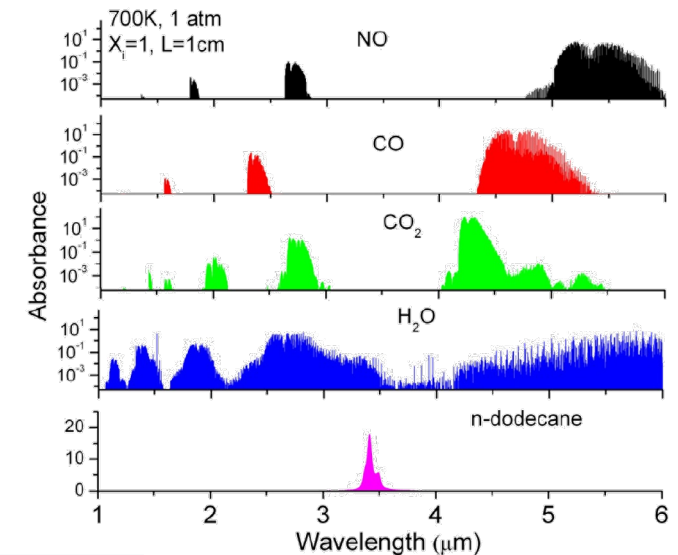
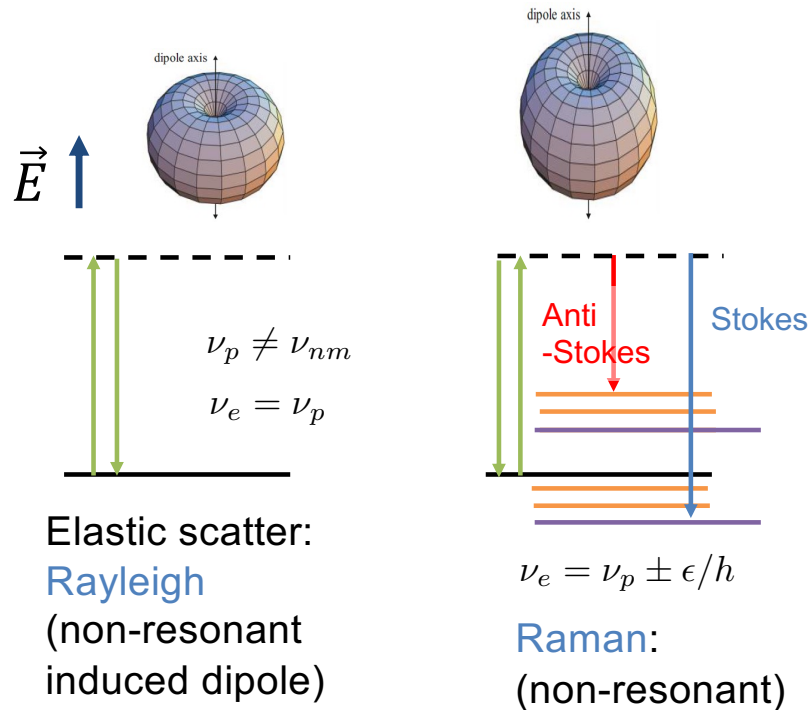
Transitions: elastic $\nu' = \nu$

inelastic $\nu' < \nu$

Radiation-molecule interactions: transitions



Hitran absorption database

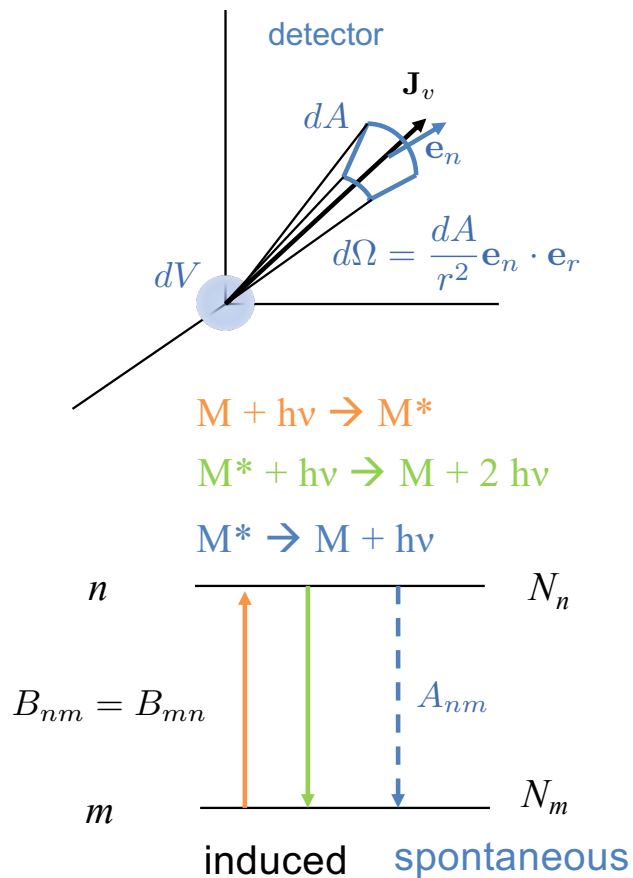


Hanson CISS 2018 Lecture 2



Output:
 Incoherent signal
 (radiates ~isotropically)
 Tricky to remove
 background noise in
 combustion conditions

Radiance and absorption



$$J_{\nu,n} = \mathbf{J}_{\nu} \cdot \mathbf{e}_n = \frac{dI_{\nu}}{d\Omega}$$

Radiative power/area/stereo radian/frequency

$$I_{\nu} = (h\nu)(\rho_{\nu}c)$$

↑
photon density (#/volume)

Energy/frequency x (photon flux) = Power/area/frequency

$$\frac{dN_n}{dt} = -N_n A_{nm} + (N_m - N_n) \rho_{\nu} B_{nm}$$

rates
population

Equilibrium:

$$\frac{N_n}{N_m} = \frac{g_n}{g_m} \exp\left(-\frac{\epsilon_{nm}}{kT}\right)$$

degeneracies

$$I_{\nu} = \frac{8\pi(h\nu)\nu^2/c^2}{\exp(h\nu/k_B T) - 1}$$

Planck's distribution

Energy levels for a rigid rotor

- Eigenvalues to the Schrödinger equation for **diatomic molecule** with fixed bond length, r_0
rotational energy levels

$$E_J = \frac{h^2}{8\pi^2 I} J(J+1), \quad J = 0, 1, 2, \dots$$

moment of inertia

$$I = \mu r_0^2$$

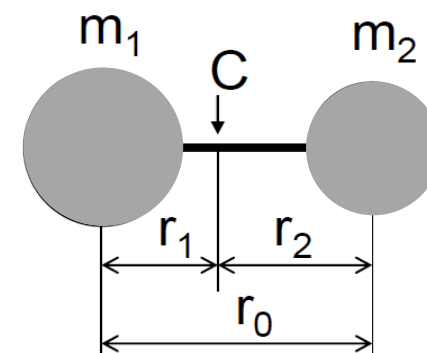
reduced mass

$$\mu = \frac{m_1 m_2}{m_1 + m_2}$$

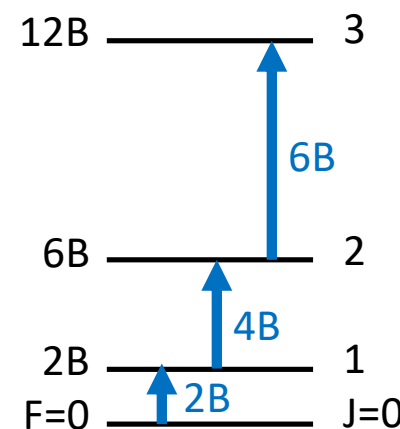
rotational energy levels in wavenumbers

$$F_J = \frac{E_J}{hc} = \frac{h}{8\pi^2 c I} J(J+1) = B J(J+1) \quad [\text{cm}^{-1}]$$

rotational constant, B



C – center of mass



Energy levels for a rigid rotor

- Selection rule, $\Delta J = \pm 1$, so for absorption $\Delta J = +1$

$$F_J = BJ(J + 1) \quad [\text{cm}^{-1}]$$

- Lines in rigid rotator absorption spectrum are evenly spaced (cm^{-1})

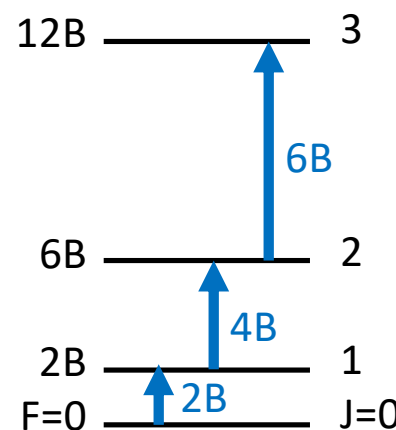
$$\bar{\nu}_{J'=1 \leftarrow J''=0} = 2B(J'' + 1)$$

- $B_{\text{CO}} \sim 2 \text{ cm}^{-1} \Rightarrow \lambda_{J'=1 \leftarrow J''=0} \sim 2.5 \text{ mm}$

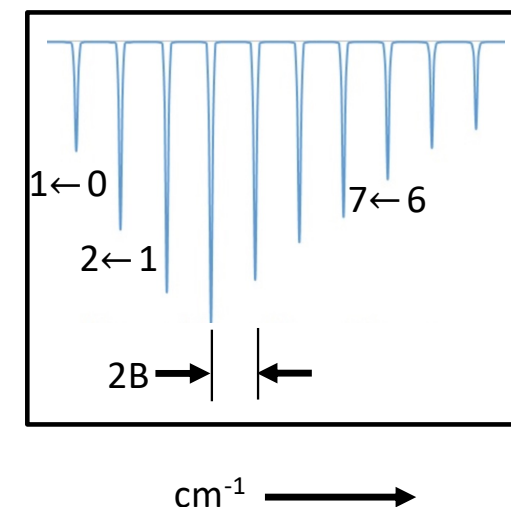
- Non-rigid rotator: add **centrifugal distortion term**,

$$F_J = BJ(J + 1) - DJ^2(J + 1)^2 \dots$$

- Interaction of light with a rotating dipole moment; only **heteronuclear** molecules have transitions



absorption spectrum
for pure rotation



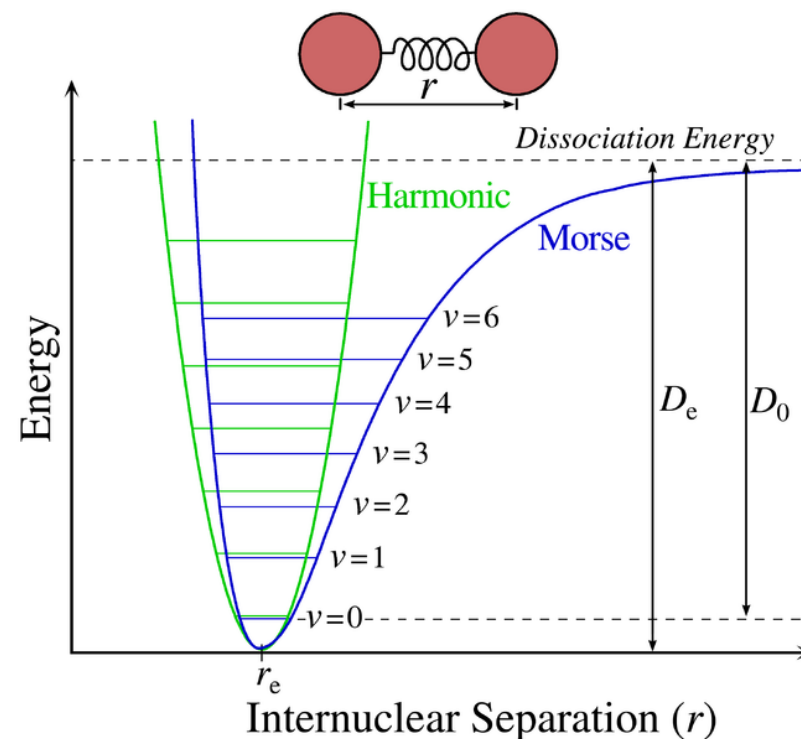
Vibrational-rotational (ro-vibrational) IR spectra

- Combine vibrational and rotational transitions (energies in the IR)
- Harmonic oscillator approximation (see CISS Linne or Hanson for details, or Google)
- Total energy (vibration plus rotation)

$$T_{v,J} = G(v) + F(J)$$

$$T_{v,J} = \omega_e(v + 1/2) - \underbrace{\omega_e x_e(v + 1/2)^2}_{\text{first anharmonic correction}} + BJ(J + 1) + \dots$$

$\omega_e = \nu/c \quad [\text{cm}^{-1}]$



http://titan.physx.u-szeged.hu/~dpiroska/atmolfiz/molecular_structure_and_spectra.html

Ro-vibrational IR spectra

- Allow non-rigid rotation, anharmonic vibration, vib-rot interaction

- Total energy (vibration plus rotation)

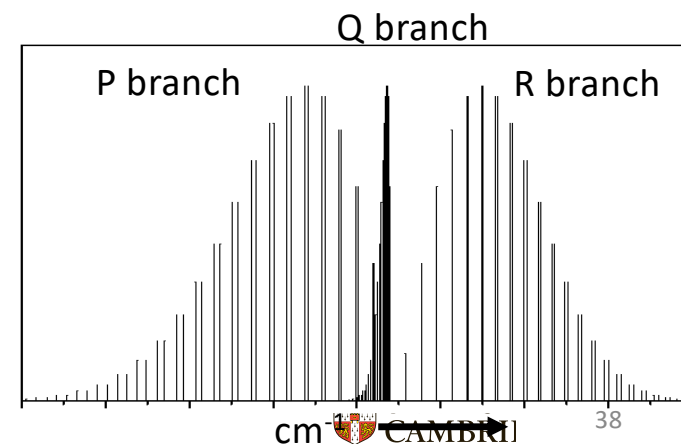
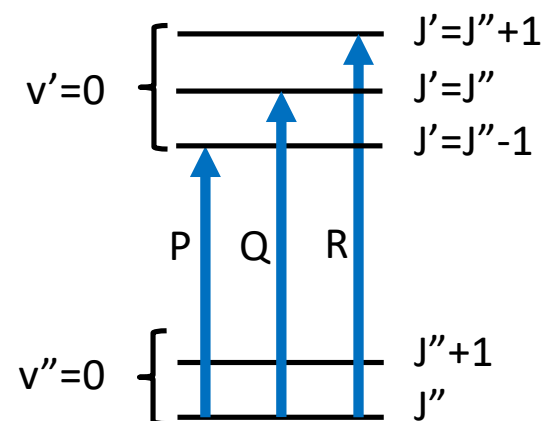
$$T_{v,J} = \underbrace{\omega_e(v + 1/2)}_{\text{simple harmonic oscillator}} - \underbrace{\omega_e x_e(v + 1/2)^2}_{\text{first anharmonic correction}} + \underbrace{BJ(J + 1)}_{\text{rigid rotator}} - \underbrace{D_v J^2(J + 1)^2}_{\text{centrifugal distortion}}$$

- Selection rule for electronic Σ state: $\Delta J = J' - J'' = \pm 1$;
Otherwise: $\Delta J = 0, \pm 1$;

- Nomenclature for branches:

Branch	O	P	Q	R	S
ΔJ	-2	-1	0	+1	+2

NO spectrum
centered near
5.35 μm ,
1875 cm^{-1}



Population distribution of internal energy states

- For a volume of gas in **thermal equilibrium**, the population distribution is given by Boltzmann statistics
- **Boltzmann distribution** function:

where:

$$f_{B,i} = \frac{N_i}{N} = \frac{g_i e^{(-E_i/k_B T)}}{\sum_j g_j e^{(-E_j/k_B T)}} \quad \text{function of } T$$

↑ $Q = \text{partition function}$

N = total number of molecules

g_i = degeneracy of level i

E_i = energy of level i

k_B = Boltzmann constant = $1.3806 \times 10^{-23} \text{ m}^2 \text{ kg s}^{-2} \text{ K}^{-1}$

Population distribution of rotational and vibrational energy states

- Internal energy: $E_{int} = E_{elec} + E_{vib} + E_{rot}$

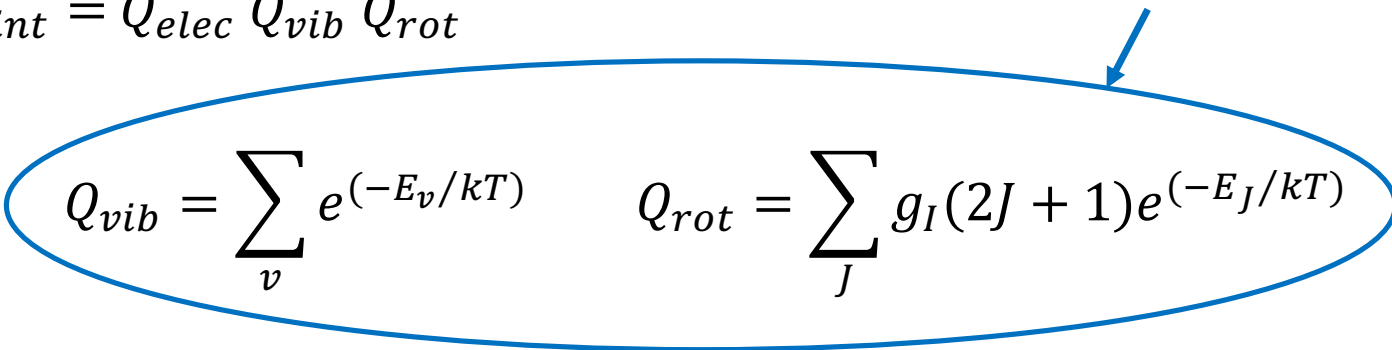
- Partition function: $Q_{int} = Q_{elec} Q_{vib} Q_{rot}$

$$Q_{elec} = \sum_n e^{(-E_n/kT)}$$

$$Q_{vib} = \sum_v e^{(-E_v/kT)}$$

$$Q_{rot} = \sum_J g_I(2J + 1)e^{(-E_J/kT)}$$

most relevant for
combustion diagnostics



- “Boltzmann fraction”

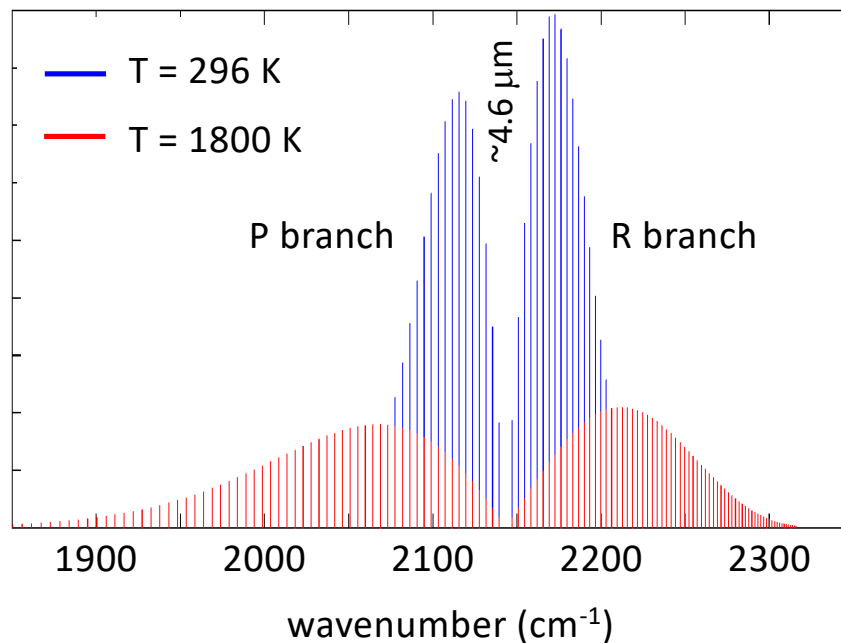
$$f_{ro-vib} = f_v f_J = \frac{e^{(-E_v/kT)}}{Q_{vib}} \frac{g_I(2J+1)e^{(-E_J/kT)}}{Q_{rot}}$$

- Equilibrium population fraction of molecules in a given ro-vibrational state before probing (ground state)

Population distribution of rotational and vibrational energy states

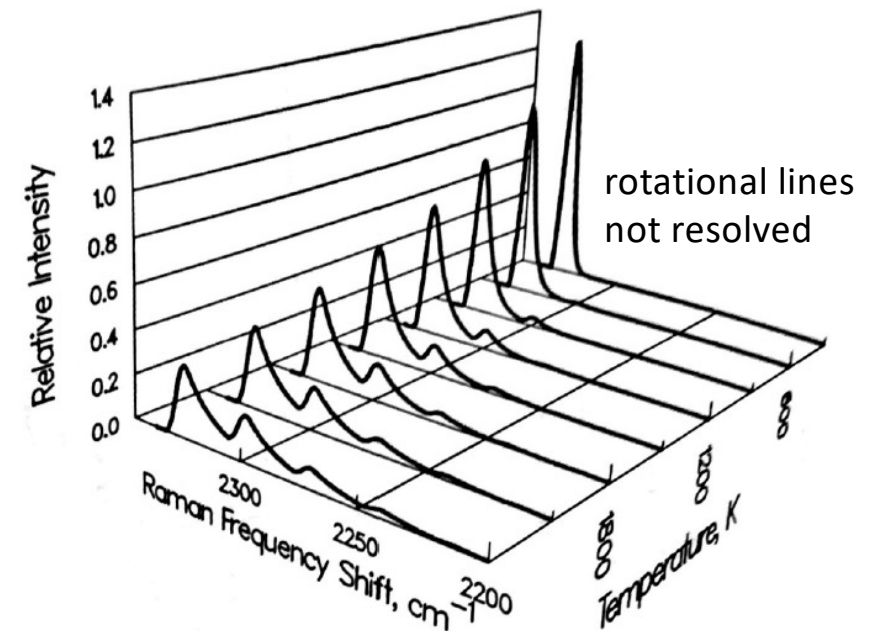
- As temperature increases, the population distribution shifts toward higher energy levels
- Basis for temperature measurements and quantitative species measurements

CO rotational line strength spectra (HITRAN)
for absorption with vibrational transition $1 \leftarrow 0$



adapted from Bo Zhou MS thesis, Lund 2011

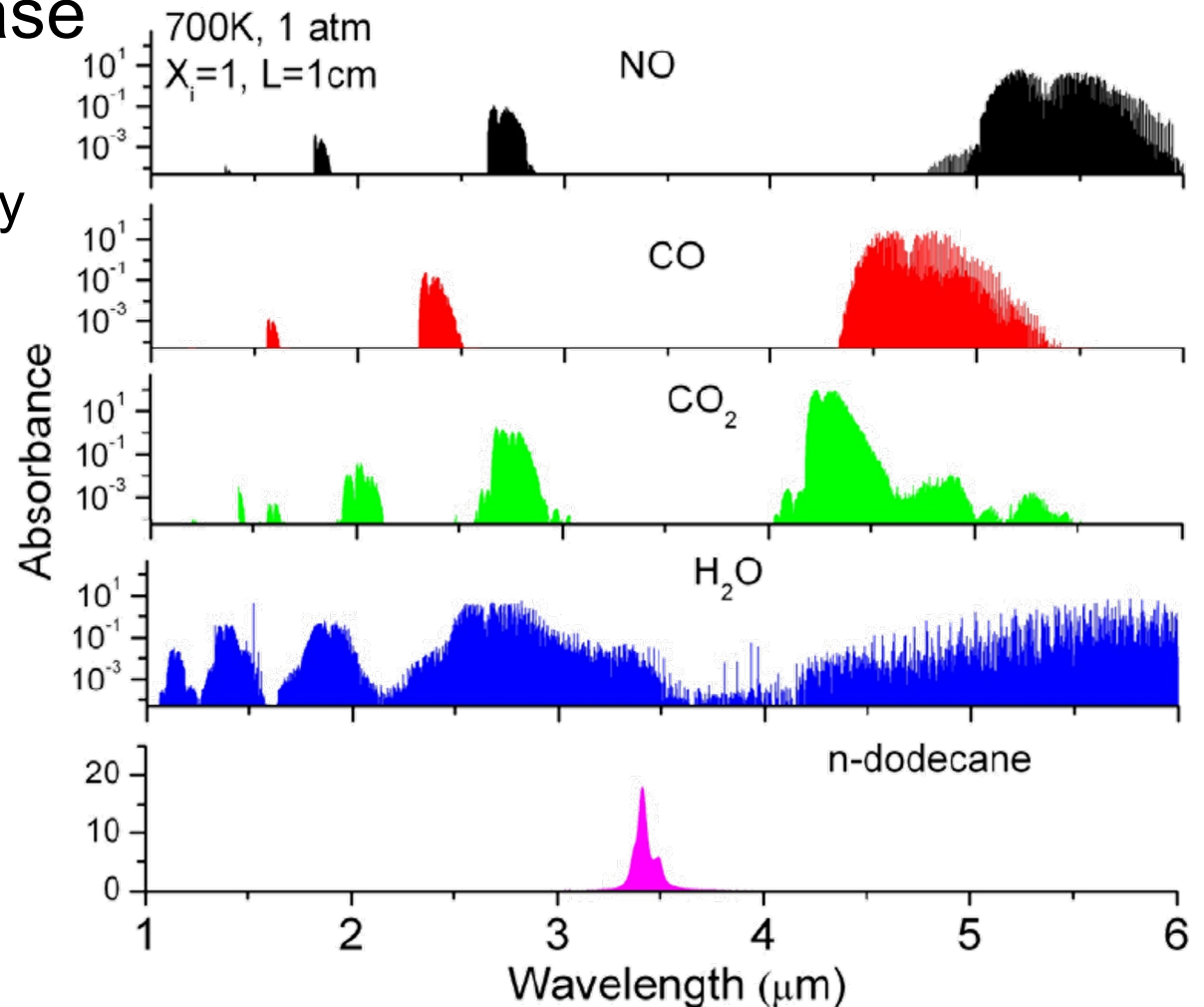
Calculated spontaneous Raman
spectra of N_2 at 300K up to 2400K



Eckbreth, Laser Diagnostics for Combustion
Temperature and Species, 1996

HITRAN/HITEMP database

- Absorption cross sections
- Resource for IR spectroscopy data on gases relevant to atmospheric science and combustion
- <https://hitran.org/>
- <https://www.spectraplot.com>



Line broadening and line shapes

Absorption/emission lines are broadened mainly by two processes

- **Doppler broadening** (thermal broadening)

- Gas molecules have a **translational energy distribution** that gets wider with increasing temperature
- Movement toward (*away from*) the laser or detector shifts the light to higher (*lower*) frequency
- Velocity distribution is Gaussian, so Doppler broadening produces a **Gaussian line function**

$$\phi_D(\nu) = \frac{2\sqrt{\ln 2}}{\sqrt{\pi}\Delta\nu_D} \exp \left[- \left(\frac{2\sqrt{\ln 2}}{\Delta\nu_D} (\nu - \nu_0) \right)^2 \right]$$

$$\Delta\nu_D \text{ (FWHM)} = 2 \sqrt{\frac{2kT \ln 2}{mc^2}} \nu_0$$

$$= 7.17 \times 10^{-7} \nu_0 \sqrt{T/M}$$

m particle mass
 M g/mole of emitter/absorber
 k Boltzmann constant
 ν_0 center frequency

- **Collisional broadening** (pressure broadening)

- Collisions between molecules perturb the radiative decay process, **shortening the lifetime of the emitter**
- Uncertainty principle $\rightarrow$ shorter time, broader line
- Time limiting process, **Lorentzian line function**:

$$\phi_C(\nu) = \frac{1}{\pi} \frac{\Delta\nu_C/2}{(\nu - \nu_0)^2 + (\Delta\nu_C/2)^2}$$

$$\Delta\nu_C \text{ (FWHM)} = 2P \sum_i X_i \gamma_{i,0} \left(\frac{T_0}{T} \right)^{n_i}$$

ν_0 center frequency
 P pressure (atm)
 X_i species mole fractions
 $\gamma_{i,0}$ broadening coeff. at T_0 ($\text{cm}^{-1} \text{ atm}^{-1}$)
 n_i temperature exponent

Line broadening and line shapes

Combine effects of collisional and Doppler broadening

- Voigt profile**

- Convolution of collisional (Lorentzian) and Doppler (Gaussian) line shape functions $a = 0$

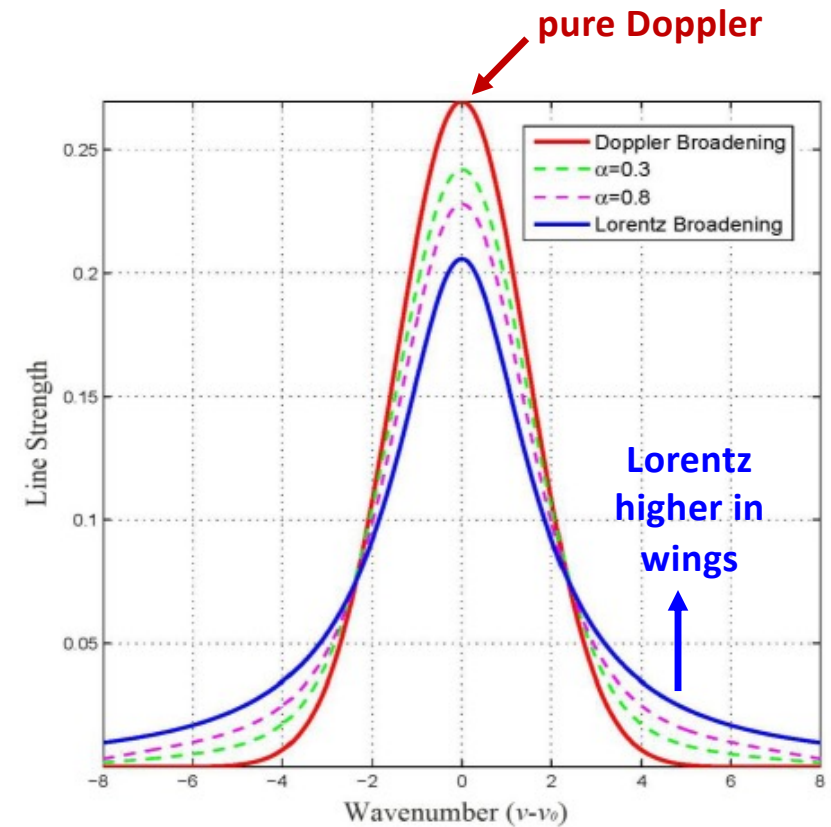
$$\phi_V(\nu) = \phi_C(\nu) * \phi_D(\nu) = \int_{-\infty}^{\infty} \phi_C(u) \phi_D(\nu - u) du$$

$$\phi_V(\nu) = \underbrace{\left\{ \frac{2\sqrt{\ln 2}}{\sqrt{\pi}\Delta\nu_D} \right\}}_{\phi_D(\nu_0)} \underbrace{\left\{ \frac{a}{\pi} \int_{-\infty}^{\infty} \frac{\exp(-y^2) dy}{a^2 + (w - y)^2} \right\}}_{\equiv V(a, w)}$$

Voigt function

$$a \cong \sqrt{\ln 2} (\Delta\nu_C / \Delta\nu_D)$$

$$w = 2\sqrt{\ln 2} (\nu - \nu_0) / \Delta\nu_D$$



Questions



Do I know what my target molecules might be?



How do I find out what the characteristic frequencies are?



What happens at high or low temperatures?



What if I only want the total number of molecules?



What if I want to measure several molecules at the same time?



Do I need to resolve the measurements spatially or in time?



Can my problem be accessed spectroscopically? Is it noisy?

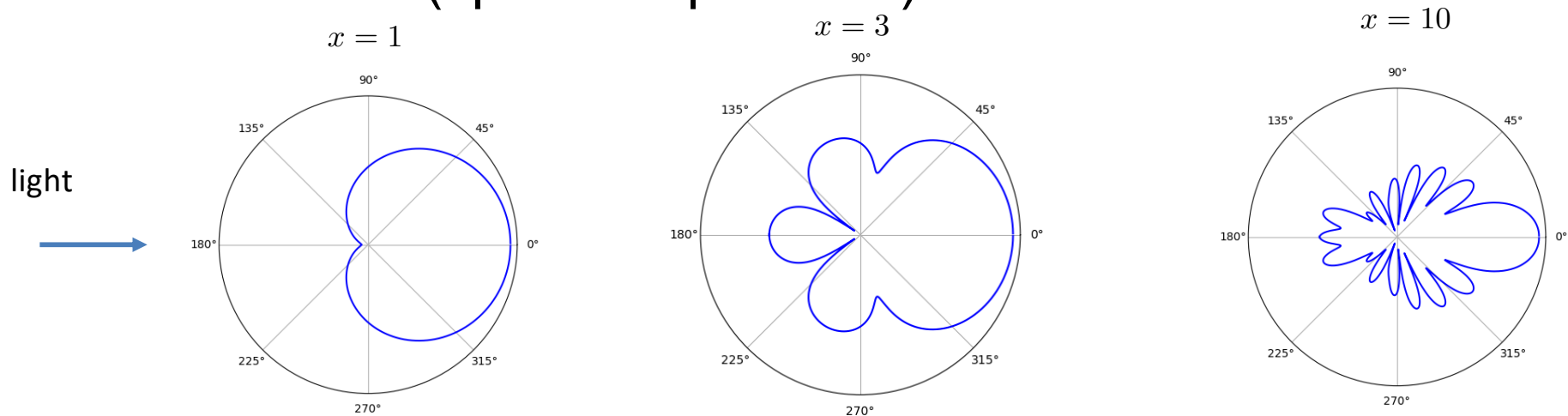


Other questions?

Outline

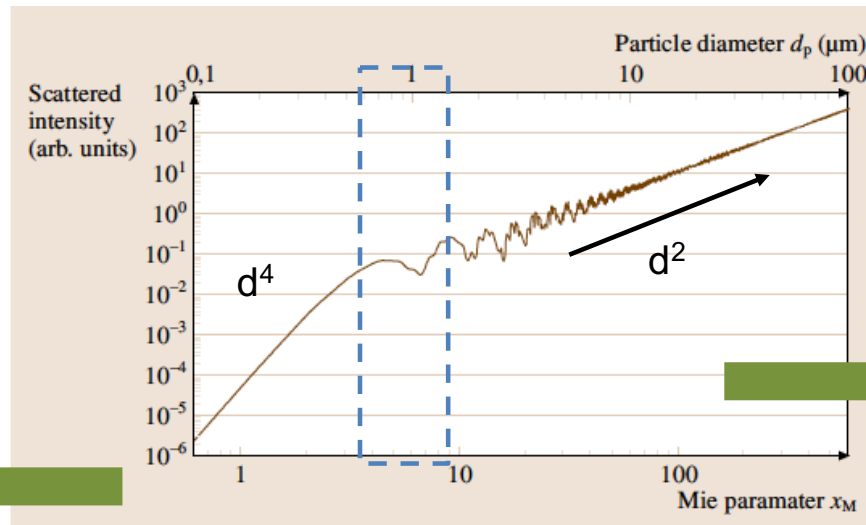
- Why and how we measure
- Fundamentals of optical diagnostics
 - Light sources and signal connection
 - Detection methods
 - Light-molecule interactions
- Incoherent
 - Scattering and particle velocimetry
 - Rayleigh scattering
 - Raman scattering
 - Absorption
 - Laser induced fluorescence
 - Laser-induced incandescence
- Coherent techniques
 - CARS
 - LIGS

Mie scatter and particle-based velocity measurements: Elastic scatter (spherical particles)



$$x = \frac{\pi a}{\lambda}$$

↑
particle radius



Measurements usually taken at 70-90 degrees forward scattering

Particles too large to follow the flow

Springer Handbook in Fluid Mechanics, 2016

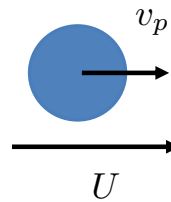
Low signal

Particle-gas velocity slip

Particle response to sudden velocity change:
What is the velocity slip ?

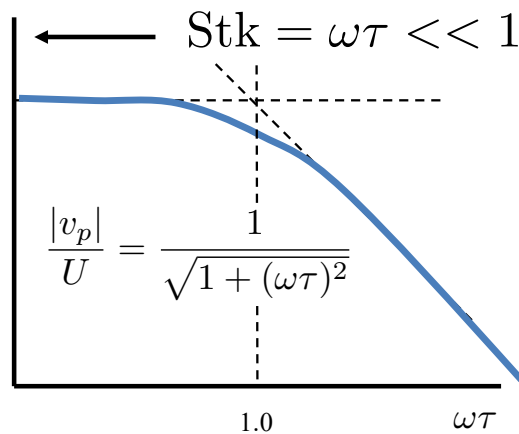
$$v_p - U = \underbrace{\frac{2}{9} \frac{a^2(\rho_p - \rho_f)}{\mu}}_{\tau} \frac{dv_p}{dt}$$

$$\tau = \frac{2}{9} \frac{a^2 \rho_p}{\mu} = \frac{\rho_p}{18\mu} d_p^2$$



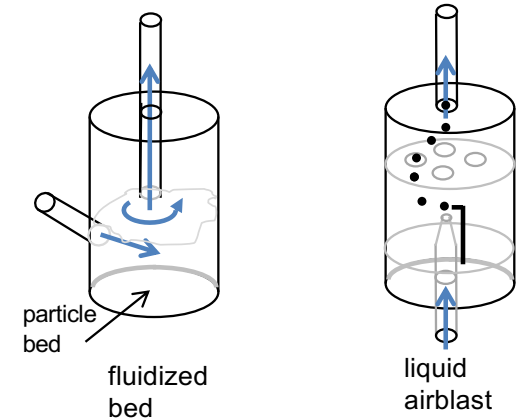
Particle seeders often have a narrow dynamic range of flow rates
1-3 x baseline value

Often tricky to get uniform particle distribution, good dispersion, etc.



Particle diameter:
small enough to follow flow, large
enough for Mie scatter

1-2 μm covers most flows at < kHz



Particle-based velocity measurements

- Seeding material for turbulent flames

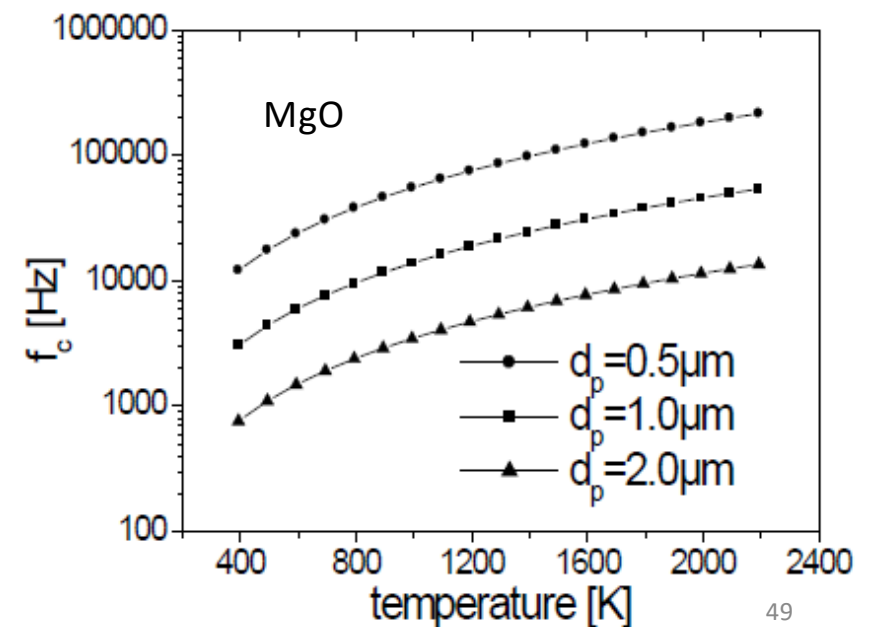
- Chemically inert
- Melting point above flame temperatures
- Small enough to follow the flow ($\sim 1 \mu\text{m}$ typ)

$$s = \left| \frac{u_f - u_p}{u_f} \right| < 1\% \quad \tau_0 = \frac{\rho_p d_p^2}{18\mu} \quad \mu = \text{dynamic viscosity}$$

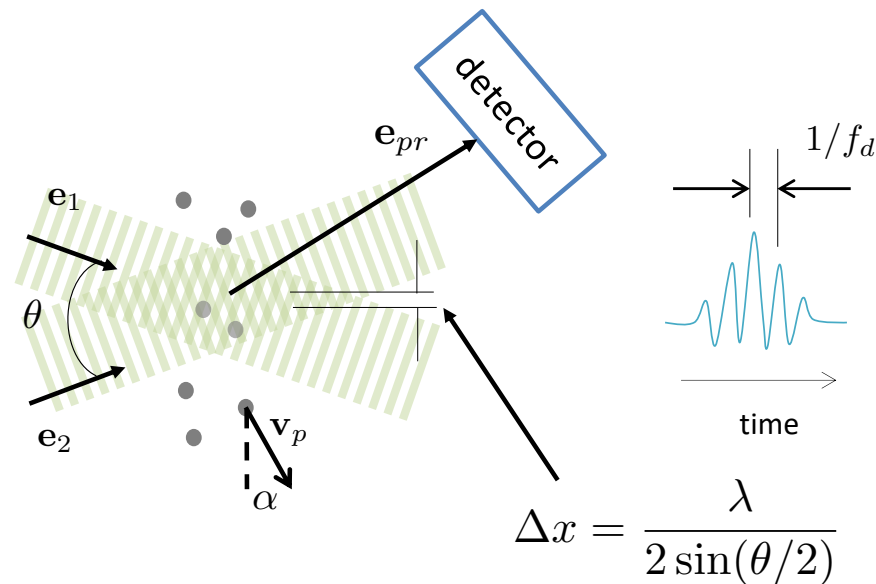
- Cut-off frequency exceeding 1% slip

$$f_c = \frac{\sqrt{(2s - s^2)}}{2\pi\tau_0 \sqrt{(1 - s^2) \left(1 + \frac{\rho_f}{2\rho_p}\right)^2 - \left(\frac{3\rho_f}{2\rho_p}\right)^2}}$$

Material	ρ_p (kg/m ³)	Melt (K)
MgO	3500	2800
ZrSiO ₄	3900-4700	2420
TiO ₂	4000	1780



Particle-based velocity measurements – laser Doppler anemometry (LDA)



One beam shifted slightly in frequency for directional sensitivity : $f_D > 0$ or < 0 (Bragg cell)

cw polarized laser
Typical powers $\sim 1-5$ W

$$f_r = f_b \frac{(1 - (\mathbf{v}_p/c) \cdot \mathbf{e}_b)}{(1 - (\mathbf{v}_p/c) \cdot \mathbf{e}_{pr})} \approx f_b + \frac{\mathbf{v}_p}{\lambda_b} \cdot (\mathbf{e}_{pr} - \mathbf{e}_b)$$

10^{14} MHz

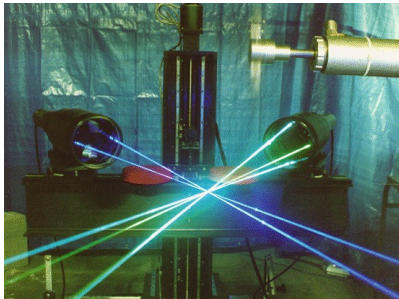
$$f_1 = f_b + \frac{\mathbf{v}_p}{\lambda_b} \cdot (\mathbf{e}_{pr} - \mathbf{e}_1)$$

$$f_2 = f_b + \frac{\mathbf{v}_p}{\lambda_b} \cdot (\mathbf{e}_{pr} - \mathbf{e}_2)$$

$$\textcircled{f_d} = f_1 - f_2 = \frac{\mathbf{v}_p}{\lambda_b} \cdot (\mathbf{e}_1 - \mathbf{e}_2) = \frac{2 \sin \theta}{\lambda_b} \textcircled{v_p} \cos \alpha$$

1 MHz

Particle-based velocity measurements – laser Doppler anemometry (LDA)



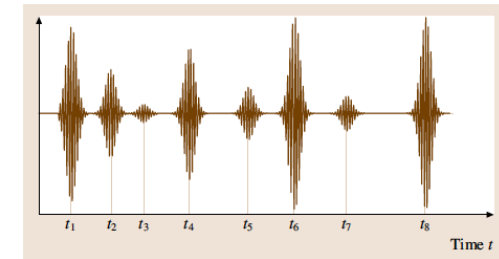
- one color per direction
- plane must cross in direction of measurement
- quasi-cw laser with $f \sim \text{MHz}$: moving fringes $> 1 \text{ W}$

Spatial resolution: typ. $200 \mu\text{m} \times 1 \text{ mm}$ (depends on crossing angle)

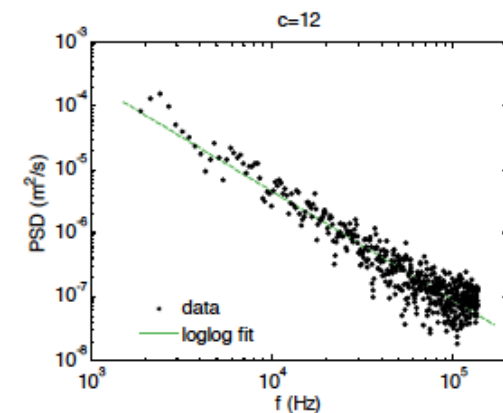
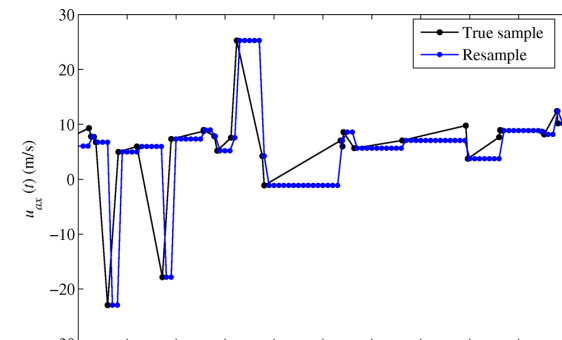
Particle sizing also possible by analyzing the phase of the scatter (PDPA).

Time resolution: depends on data rate = function of seeding, but typically **10 kHz** possible

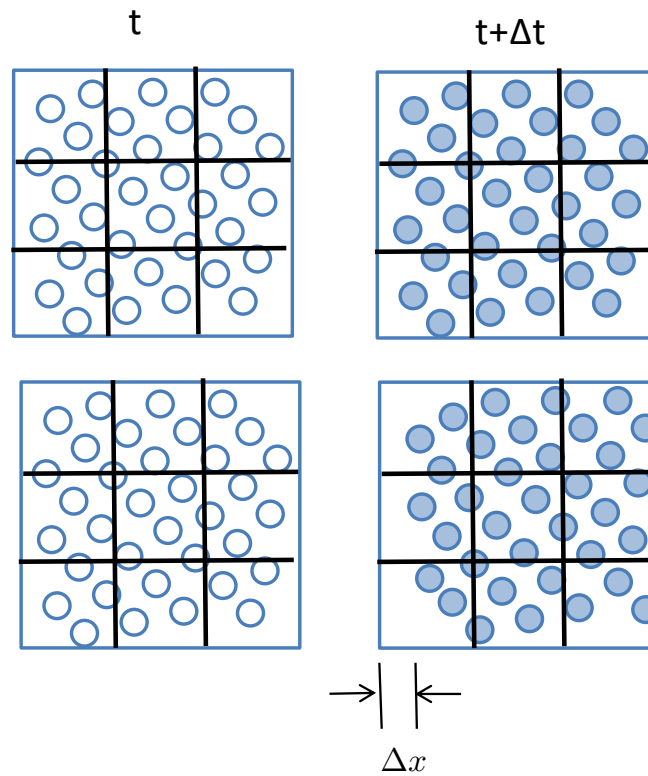
bursts



averaging and resampling



Particle image velocimetry

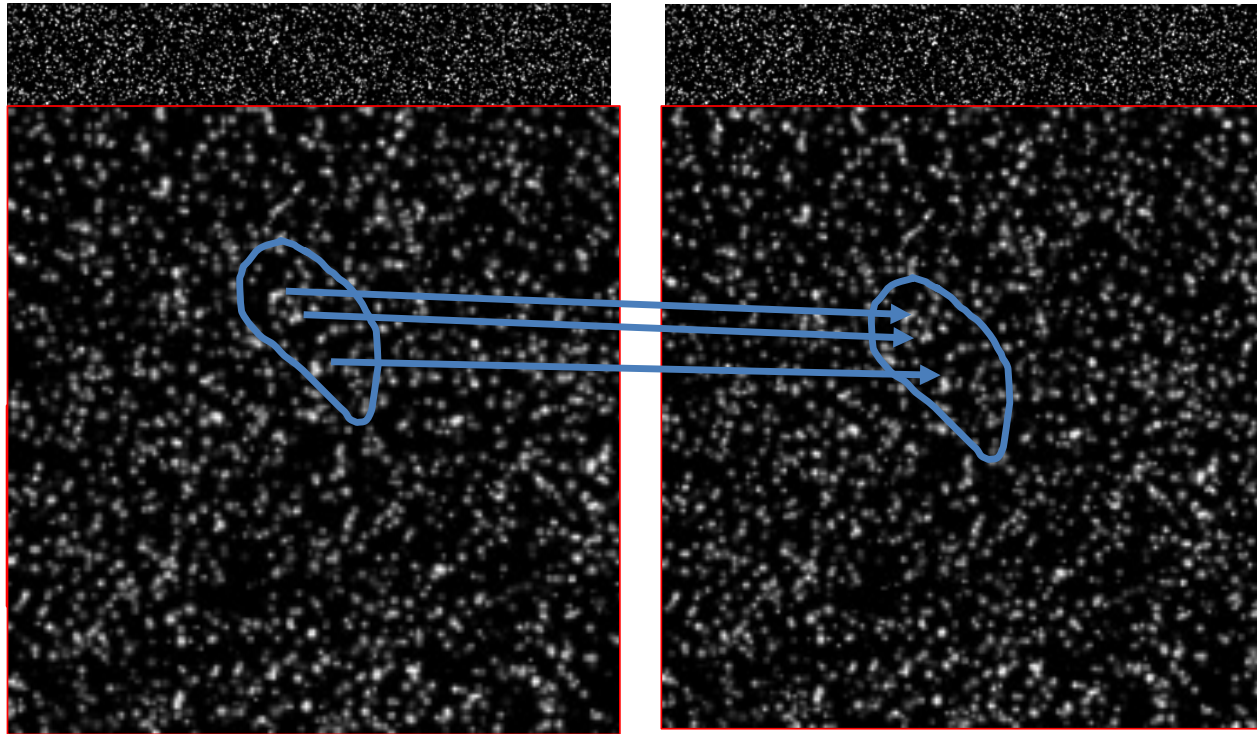


$$\Delta x = 0$$

$$u = 0$$

$$u = \frac{\Delta x}{\Delta t}$$

Sample PIV images: zooming in

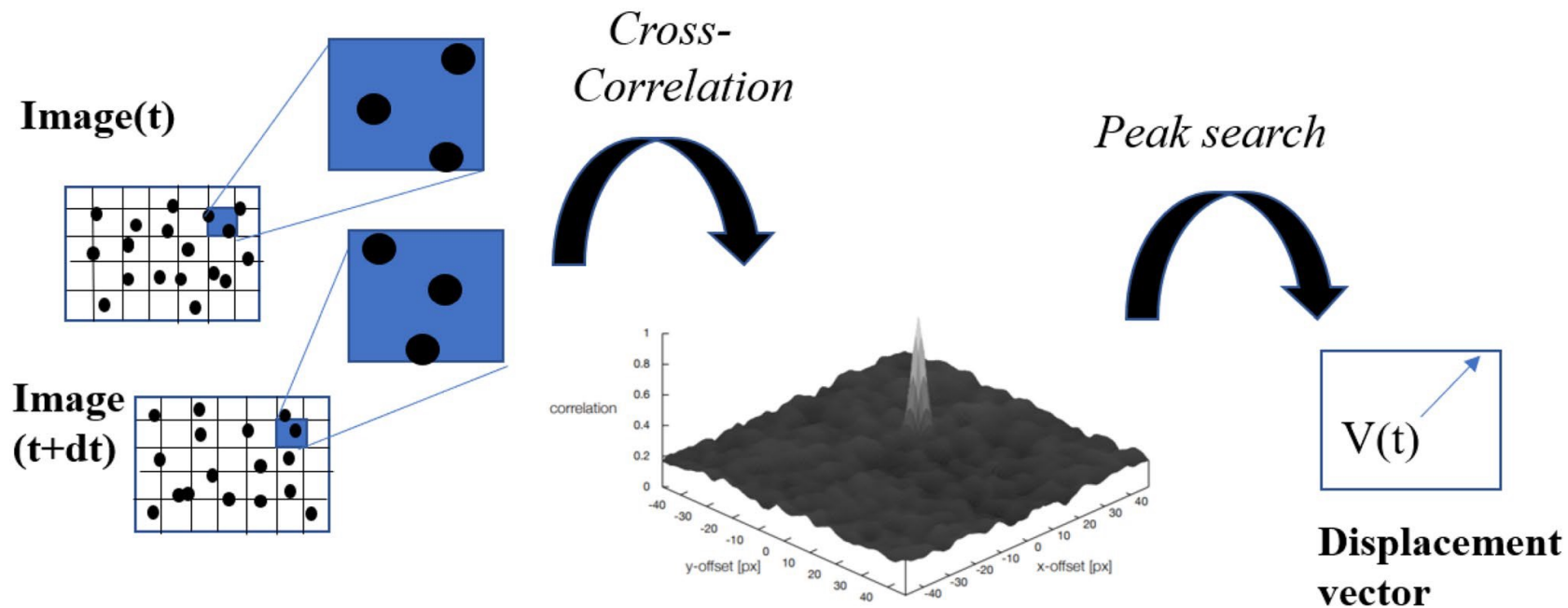


pivchallenge.org

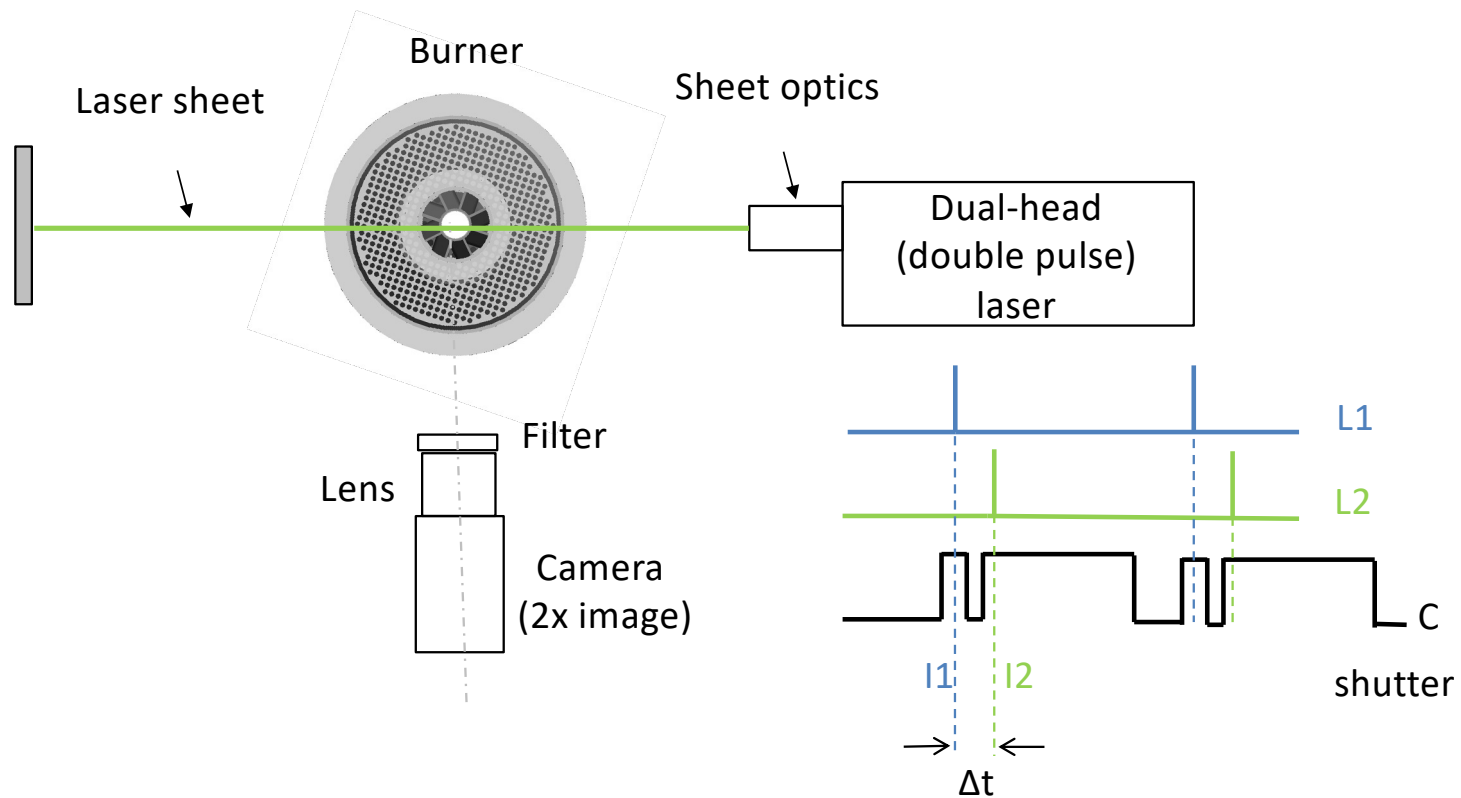
Correlation

$$R(\mathbf{d}) = \frac{1}{\|\mathcal{W}(\mathbf{x})\|} \sum_{\mathbf{y} \in \mathcal{W}(\mathbf{x})} I_1(\mathbf{y}) \times I_1(\mathbf{y} + \mathbf{d}(\mathbf{x}))$$

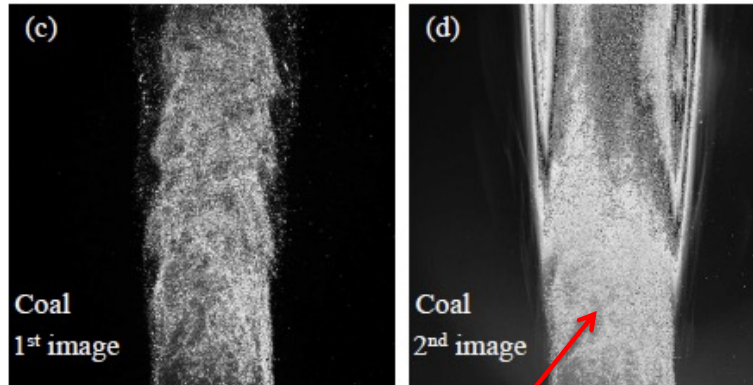
FFT algorithm for cross correlation (CC)



Typical layout: 2D PIV

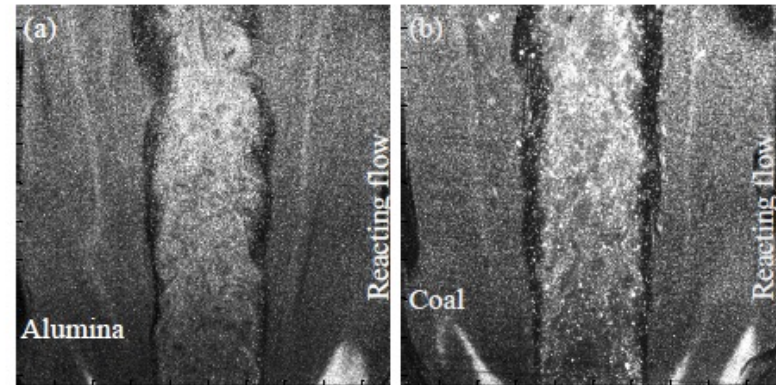


Luminosity, exposure time, interval



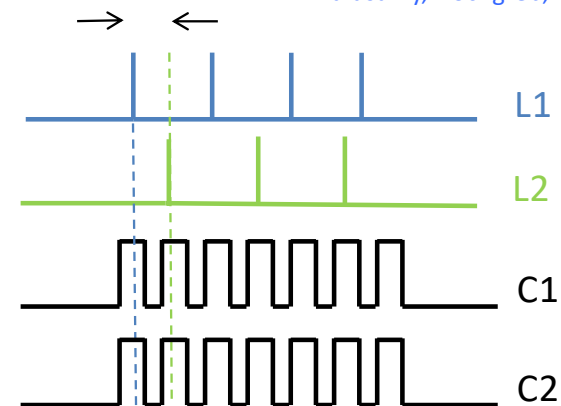
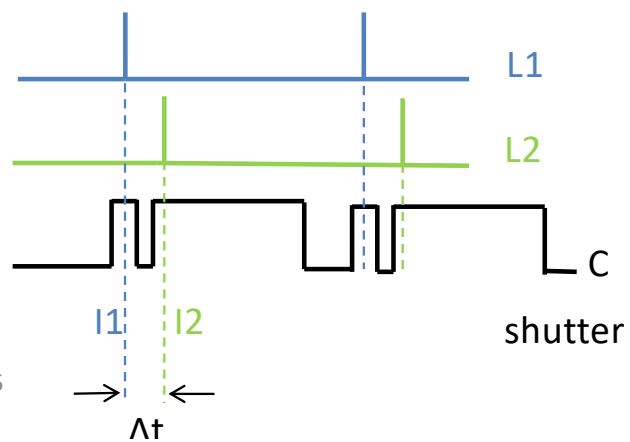
Low speed PIV

Overexposure

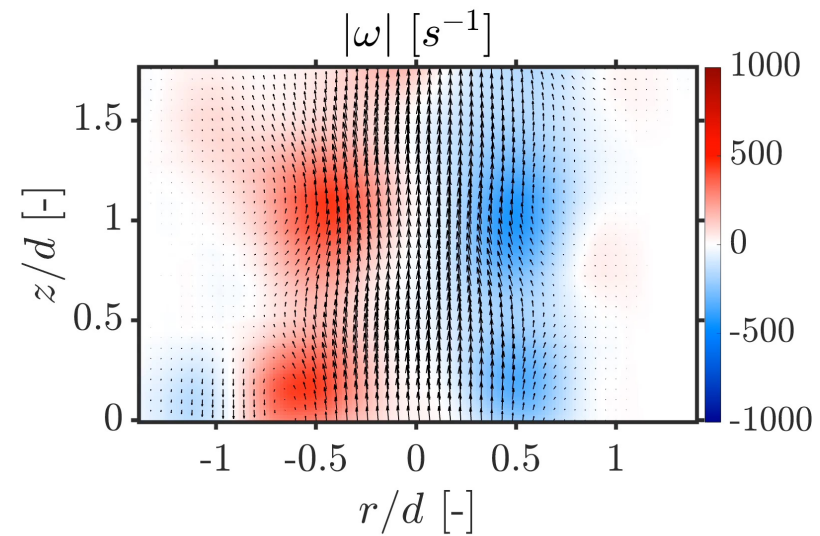
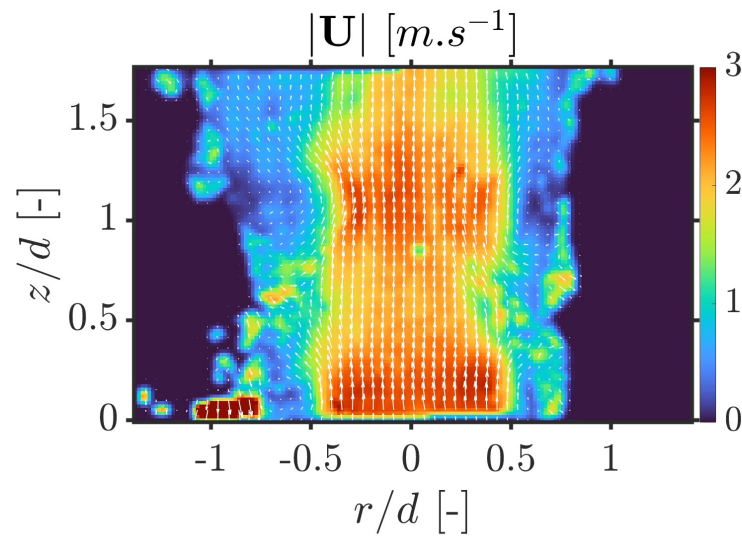


High frequency PIV (short exposure)

Balusamy, Hochgreb, EIF 2013

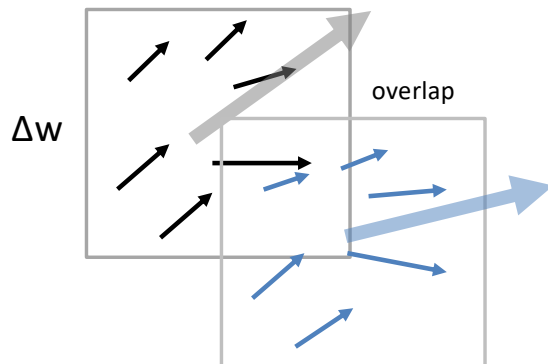


PIV sample images

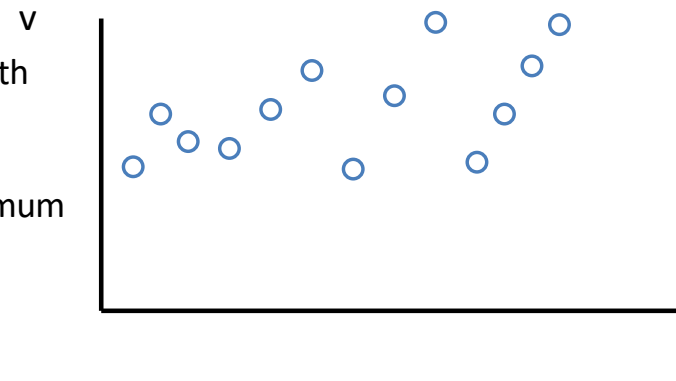
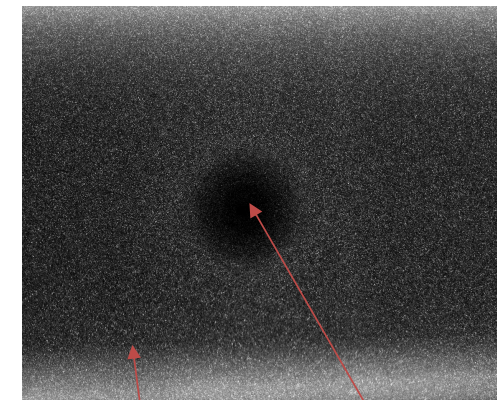


PIV parameters and limitations

- **Spatial resolution for raw images:** max. 4 x 4 cm @ 20 mJ @ 2k x 2k cameras: $< \sim 20 \mu\text{m}/\text{px}$
- **Seeding:** optimal typ. 4-6 particles per interrogation volume
- **Interrogation volume:** typ $\sim 16\text{-}32 \text{ px}$: $\Delta w = 32 \text{ px} = 0.6 \text{ mm}$, with overlap.



- **Time resolution:** max $f=10 \text{ kHz}$ (signal typ. decreases with frequency)
- **Time interval:** $\Delta x \sim v / \Delta t \sim \Delta w \rightarrow$ limits max velocity
HS PIV: multiple images can be used to interpolate optimum min velocity for high dynamic range



HS PIV vs LDA

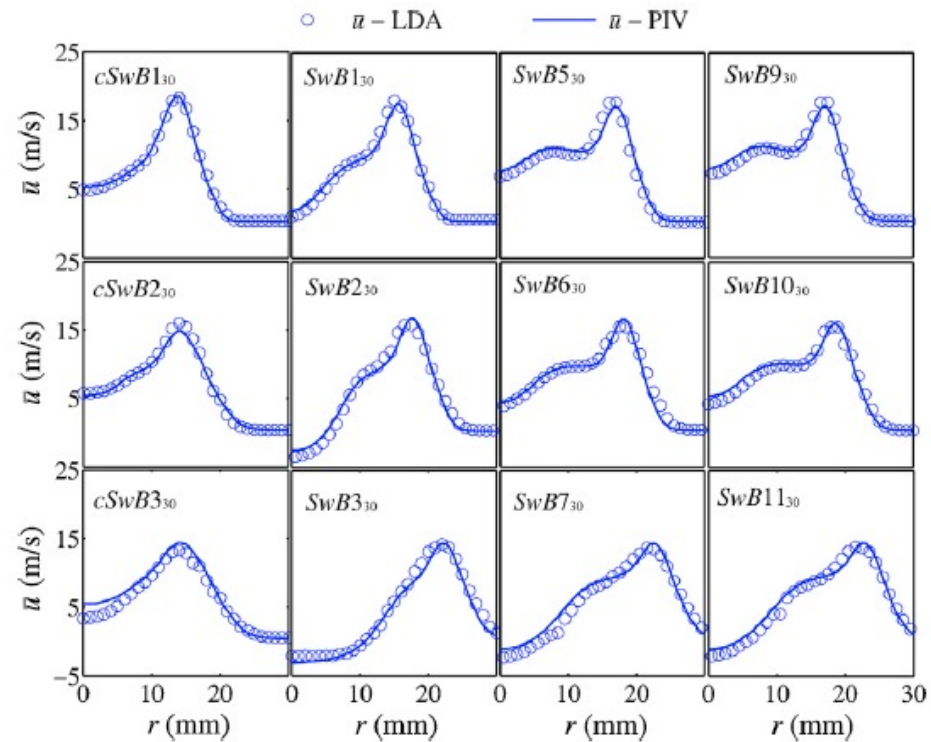
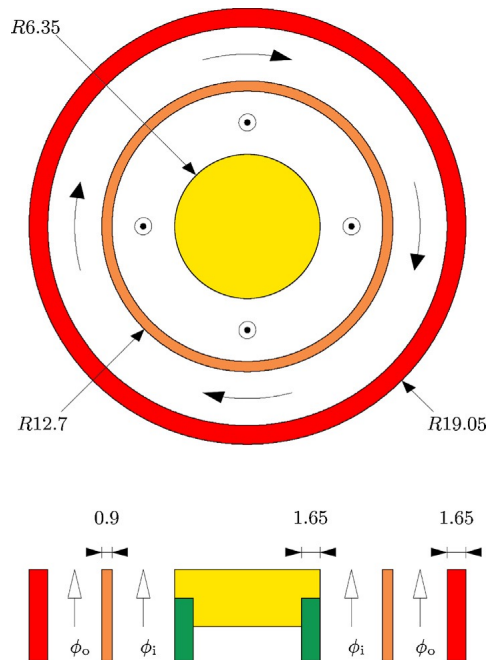
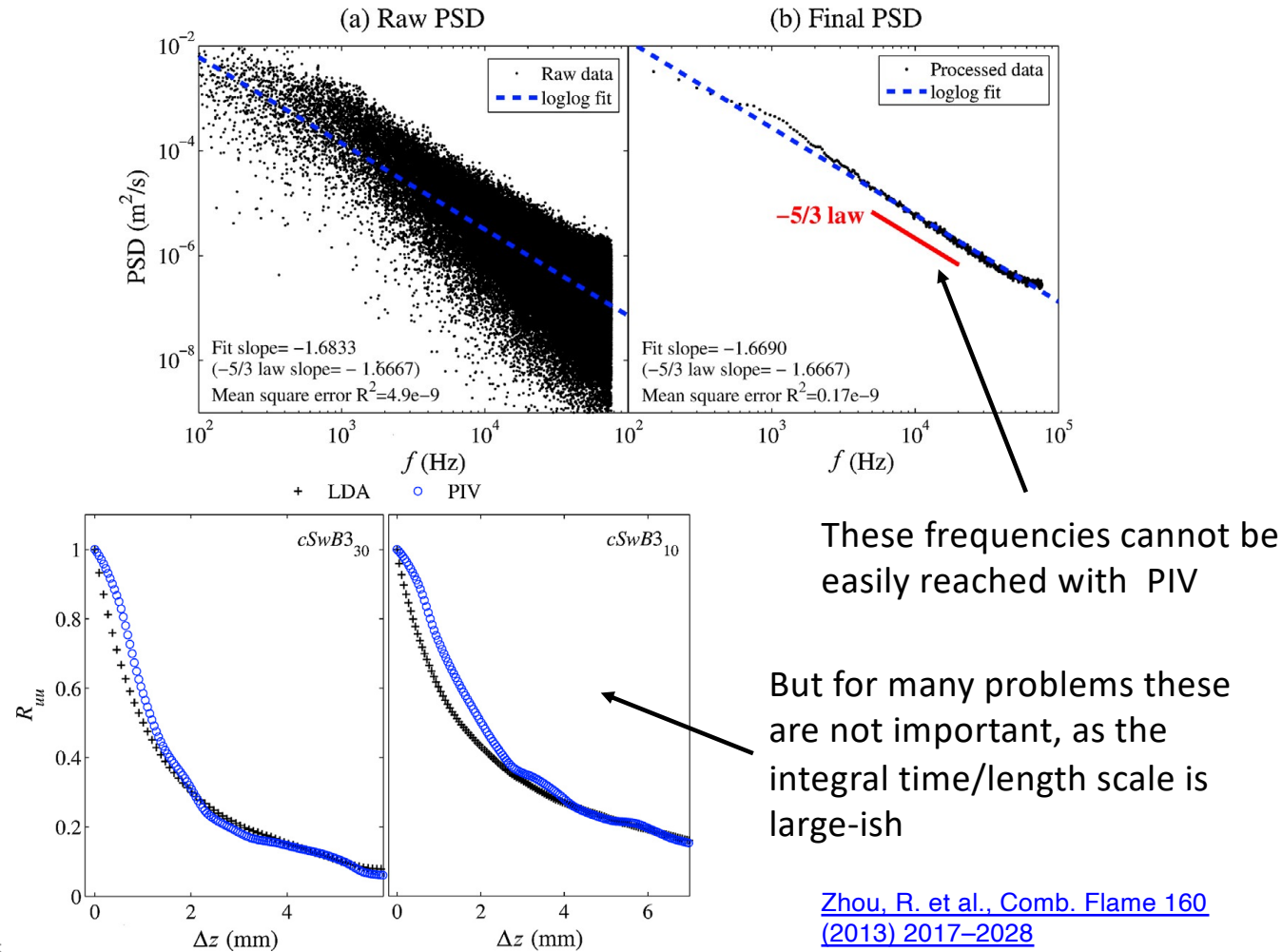


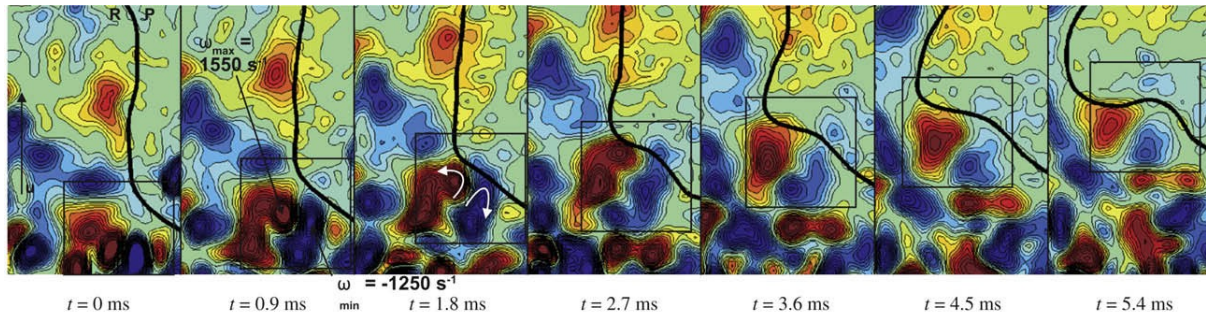
Fig. 7. Radial profiles of mean axial velocity for all non-reacting and reacting cases, extracted at $z = 30$ mm from PIV and LDA.

[Zhou, R. et al., Comb. Flame 160 \(2013\) 2017–2028](#)

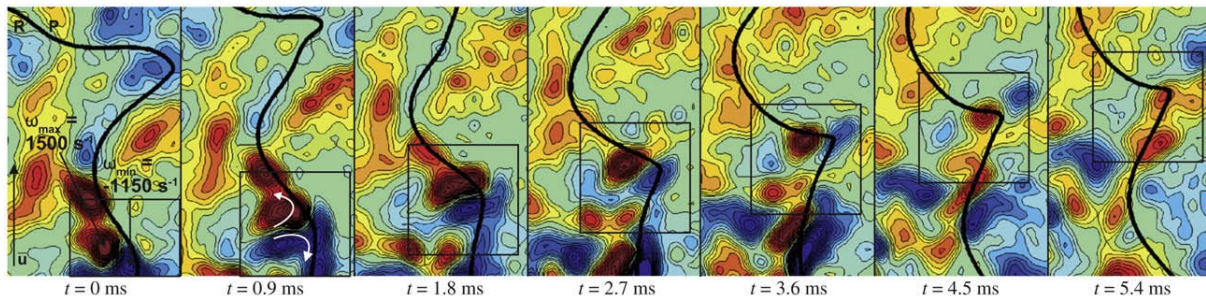
LDA dynamic range



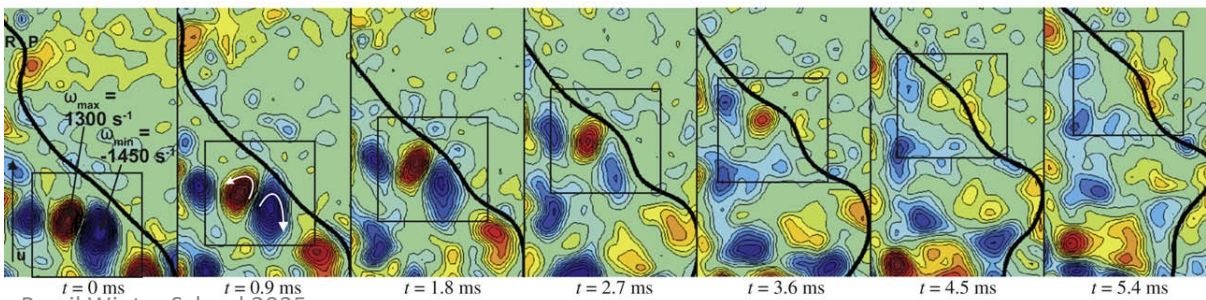
PIV



(a) A moderate amplitude wrinkle is formed



(b) A larger amplitude wrinkle is formed



(c) No wrinkle is formed

Follow vorticity across the flame

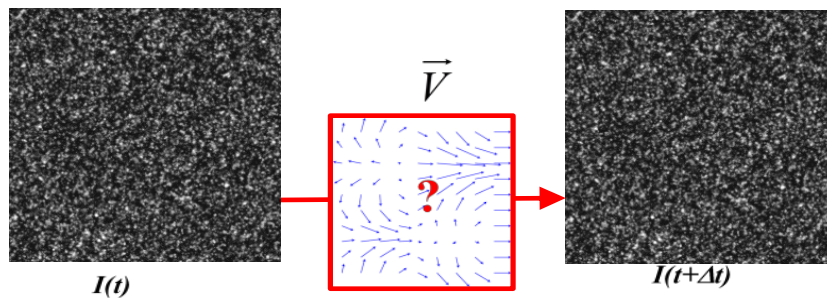
[Steinberg, A. and Driscoll, J.
 Combustion and Flame 156 \(2009\) 2285–2306](#)

Optical Flow Velocimetry (OFV)

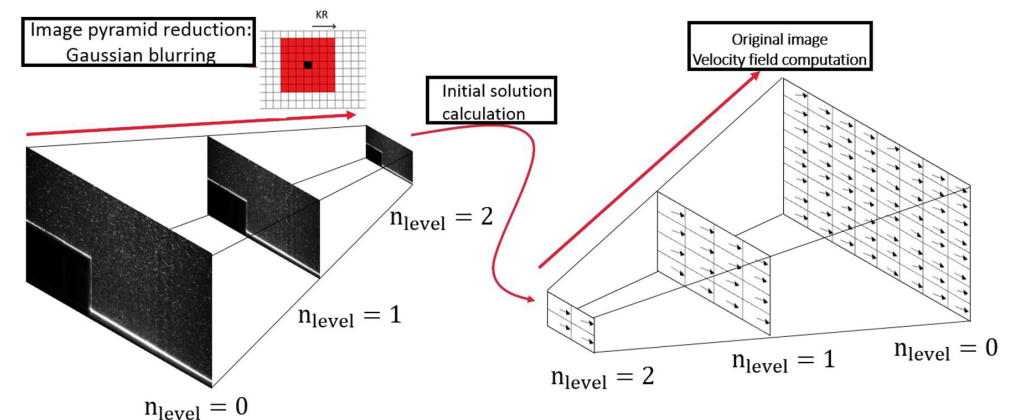
- The velocity field is estimated from the DFD equation by forming a minimization problem

$$\hat{\underline{v}} = \underset{\underline{v}}{\operatorname{argmin}} J_D(I_0, I_1, \underline{v}) + \lambda J_R(\underline{v})$$

- Goal: Find the velocity field that transforms particle image $I(t)$ to $I(t+\Delta t)$



- Hierarchical 'pyramid' algorithms GPU optimised

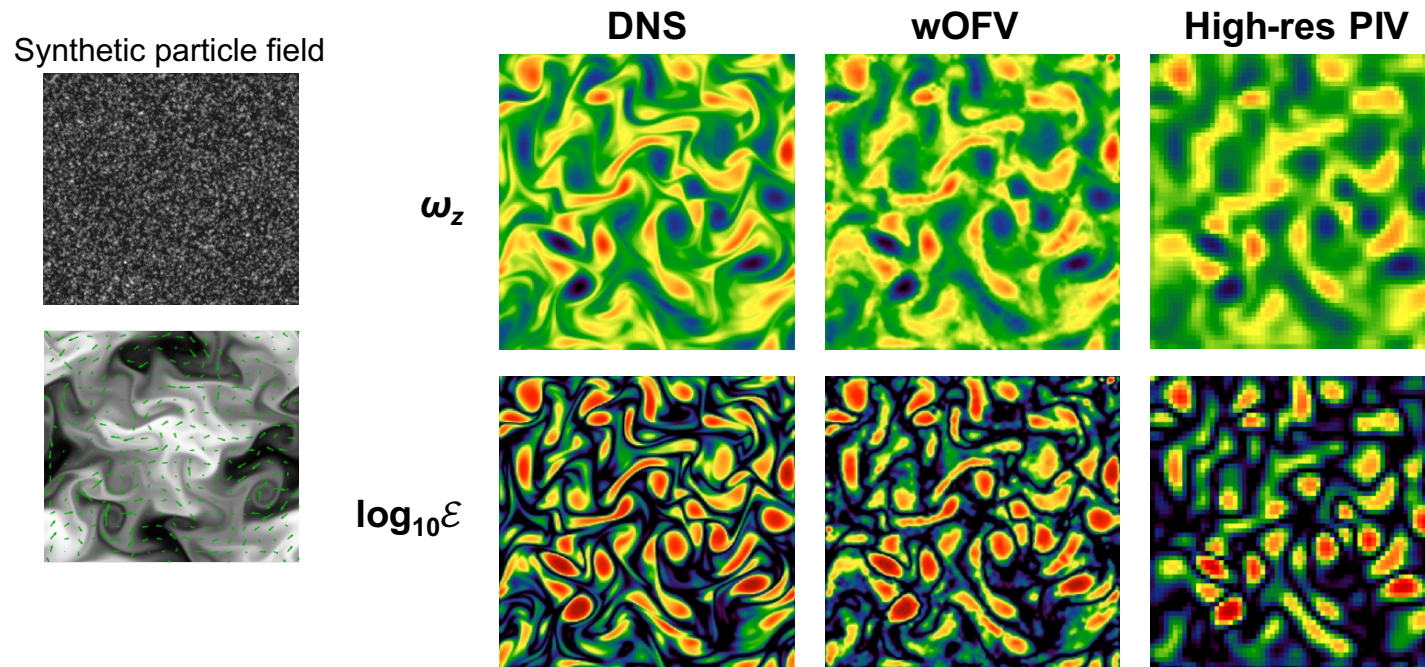


Tradeoffs between resolution, computational effort and accuracy
Different problems may be more effectively tackled by different algorithms

Wavelet-Based Optical Flow Velocimetry (wOFV)

wOFV slides from
Jeff Sutton, OSU

- Test case: 2D DNS ($Sc = 0.7$, $Re = 3000$ isotropic turbulence)^{2,4}

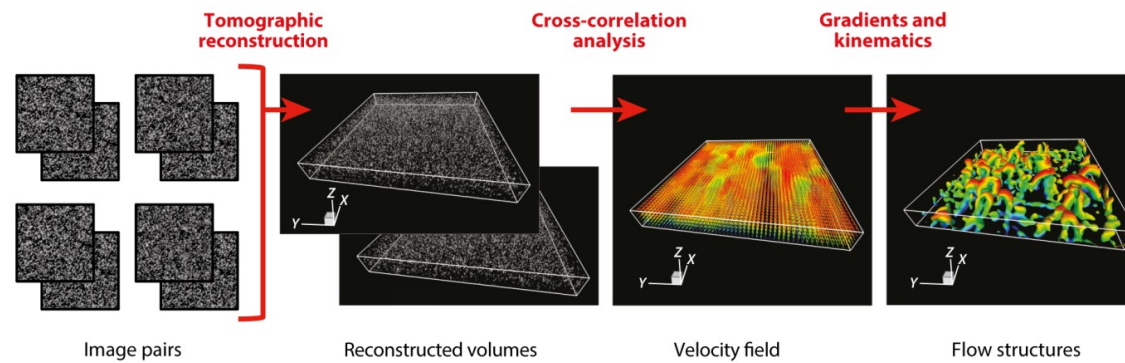
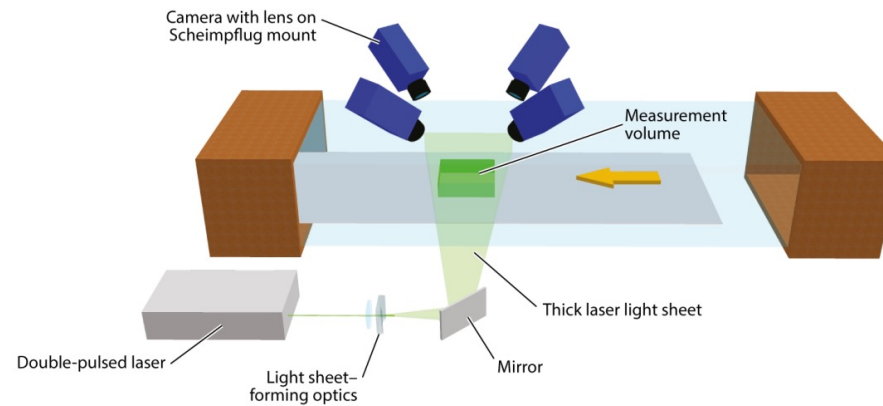


- Velocity accuracy is greatly improved; error is $1/4^{\text{th}}$ that of correlation-based PIV
- Resolution can be significantly improved

² Schmidt, B. E., Sutton, J.A., *Experiments in Fluids* 60.3 (2019): 37.

⁴ Schmidt, B.E., Sutton, J.A., "Improvements in the accuracy of wavelet-based optical flow velocimetry using an efficient and physically based implementation of velocity regularization", to be submitted, *Experiments in Fluids*, 2019.

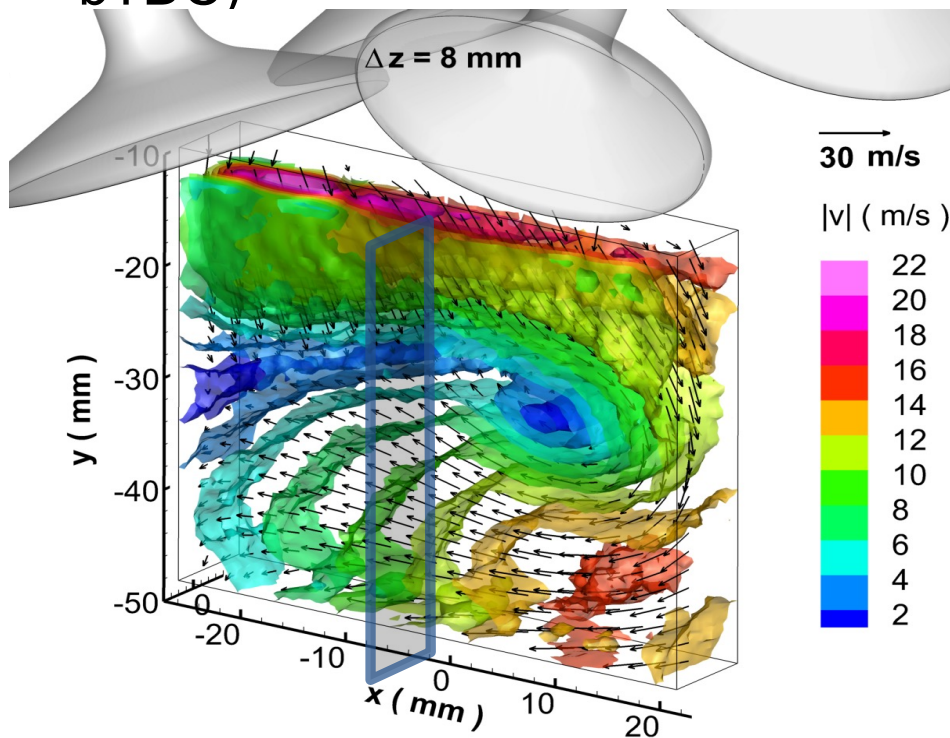
Tomographic PIV



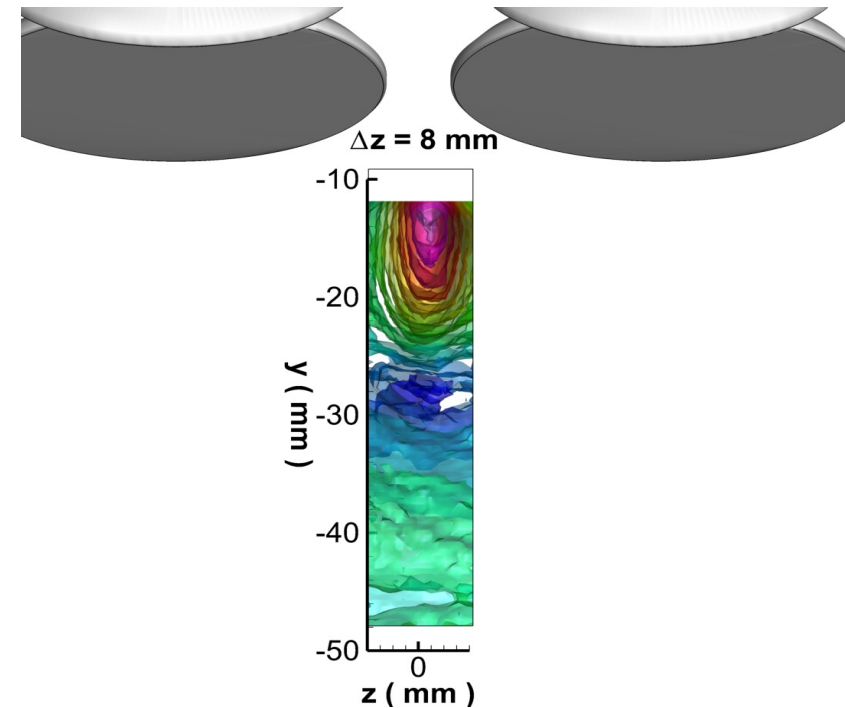
 Westerweel J, et al. 2013.
Annu. Rev. Fluid Mech. 45:409–36

Application example: Tomo-PIV of in-cylinder engine flow

- Iso-surface of average velocity magnitude during intake stroke (270° bTDC)

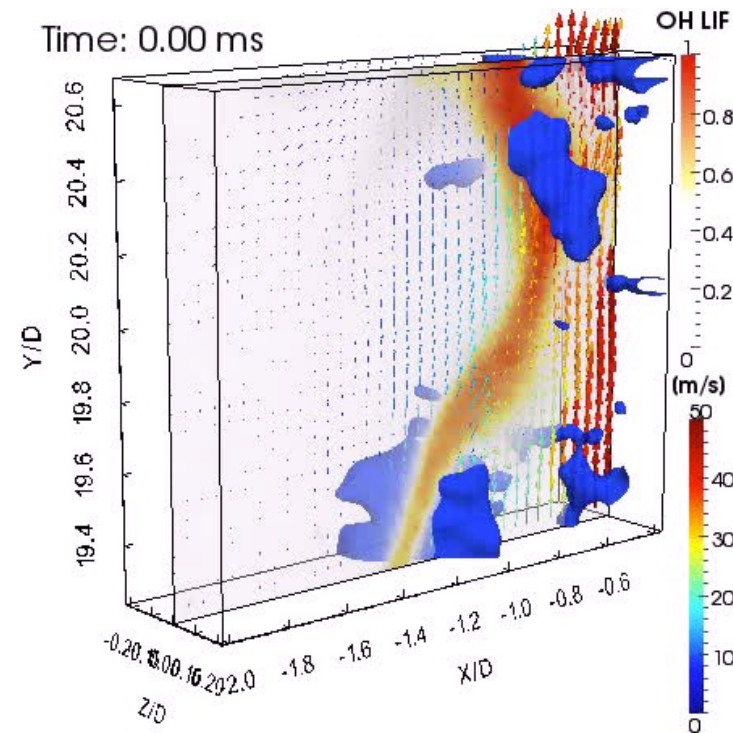


Piston at -51mm



Average over 300 cycles

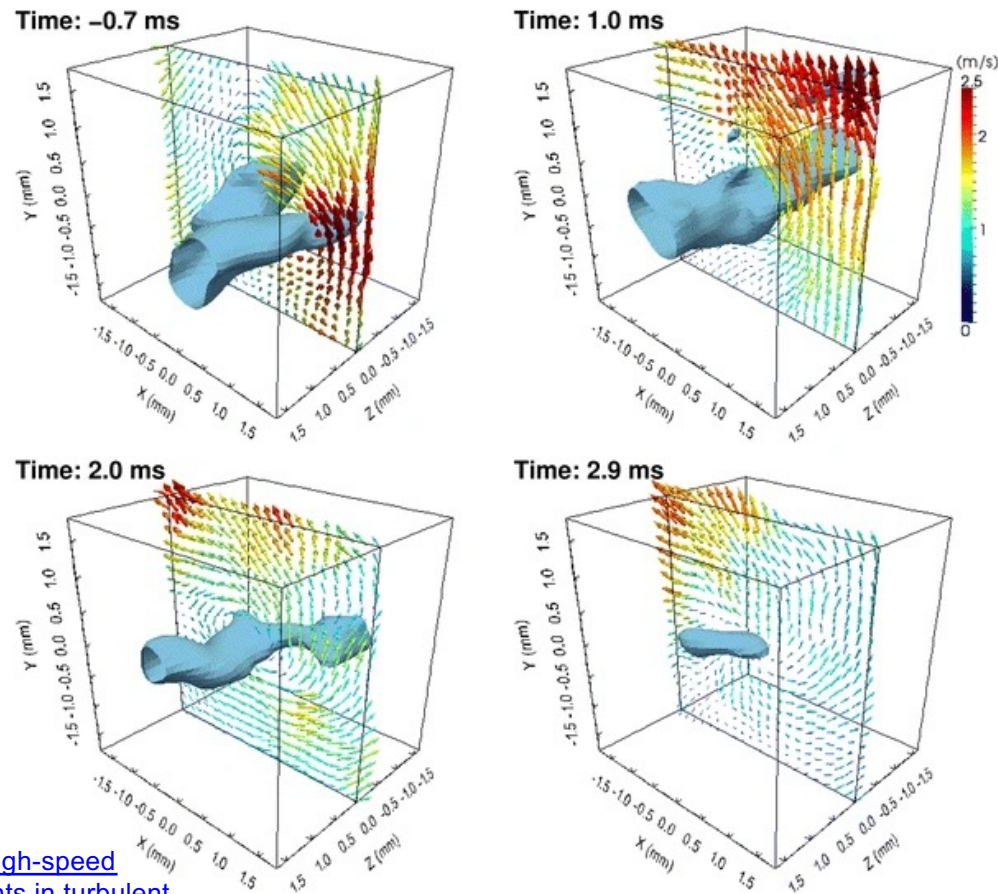
Tomographic PIV and planar OH PLIF



[Coriton, B., Steinberg, A.M. & Frank, J.H. High-speed tomographic PIV and OH PLIF measurements in turbulent reactive flows. *Exp Fluids* 55, 1743 \(2014\).](#)

Simultaneous 10-kHz TPIV and OH PLIF measurements obtained in the stabilization region of a lifted jet flame. Blue isosurfaces correspond to an enstrophy of $\omega^2 = 15 \times 10^6 \text{ s}^{-2}$. Velocity vectors are represented in the same plane as the OH PLIF images (1 out of 16 in-plane vectors displayed)

Tomographic PIV and planar OH PLIF

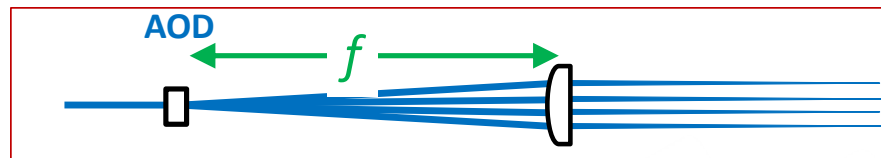


[Coriton, B., Steinberg, A.M. & Frank, J.H. High-speed tomographic PIV and OH PLIF measurements in turbulent reactive flows. *Exp Fluids* 55, 1743 \(2014\).](#)

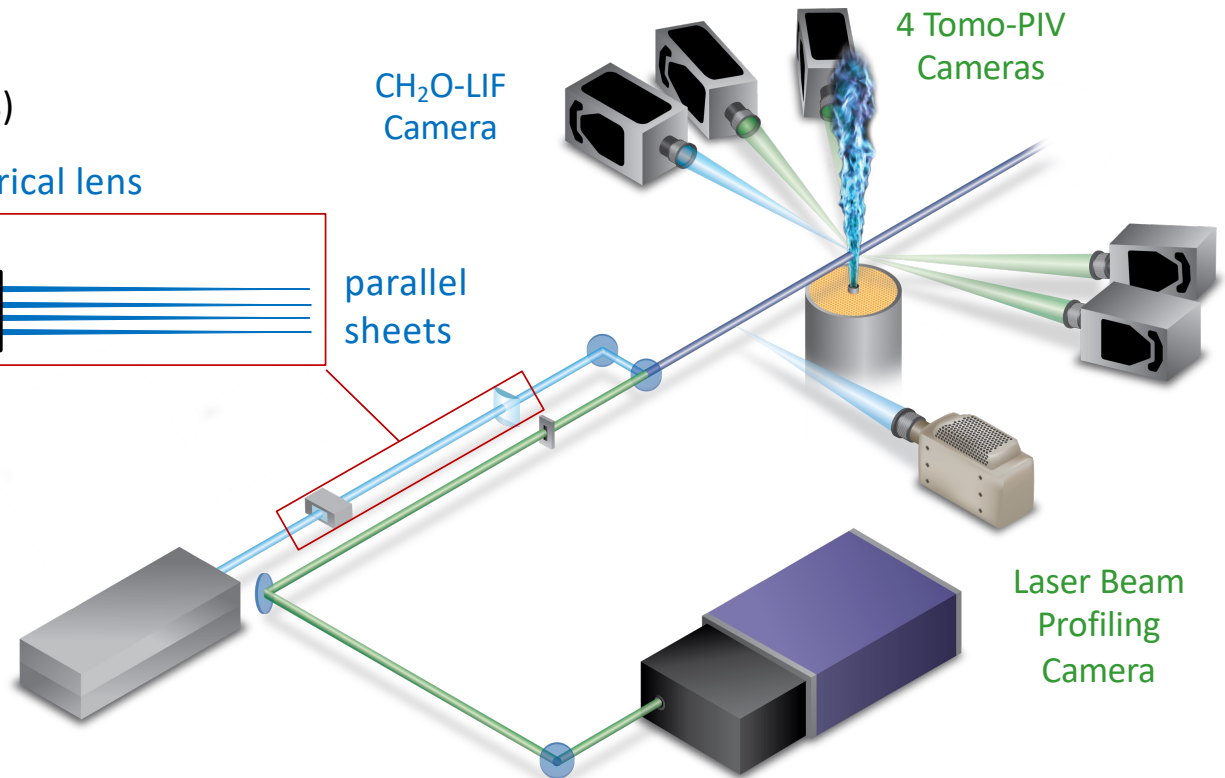
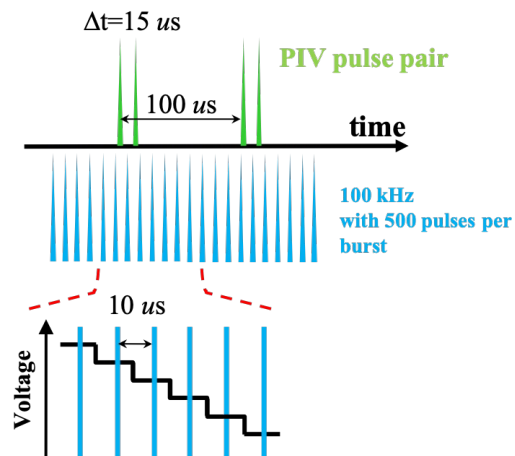
High-Speed 3D LIF and Velocity Measurement Capability

Raster scanning of laser sheet
10 kHz duty cycle (1 sweep = 100 μ s)

acousto-optic deflector cylindrical lens



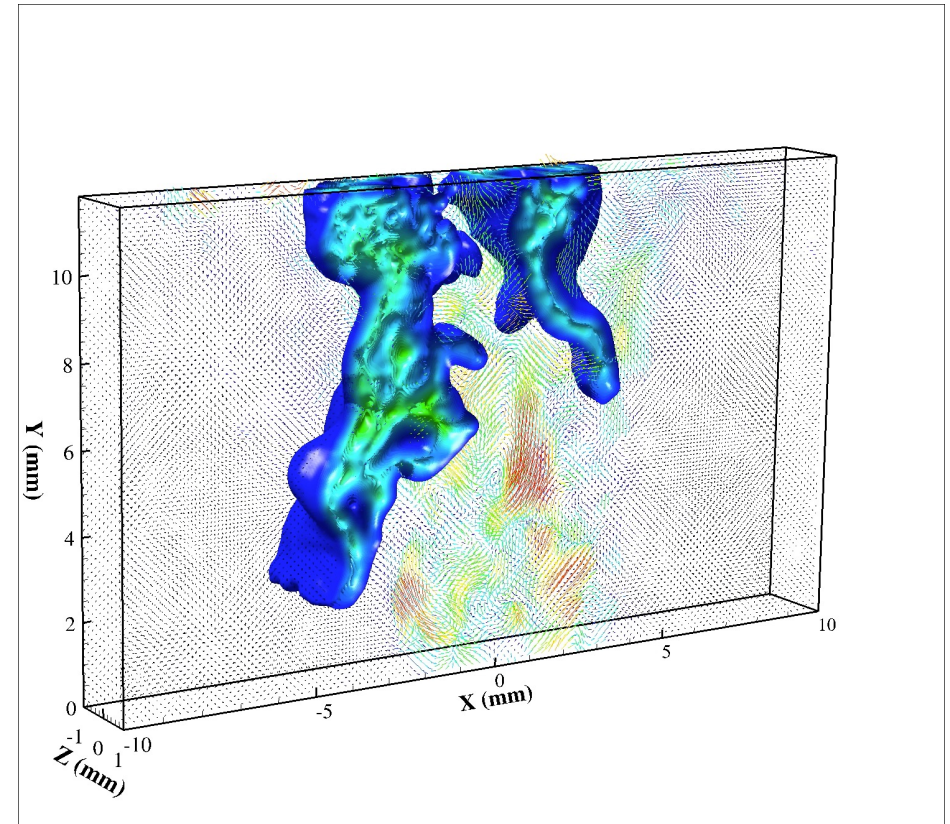
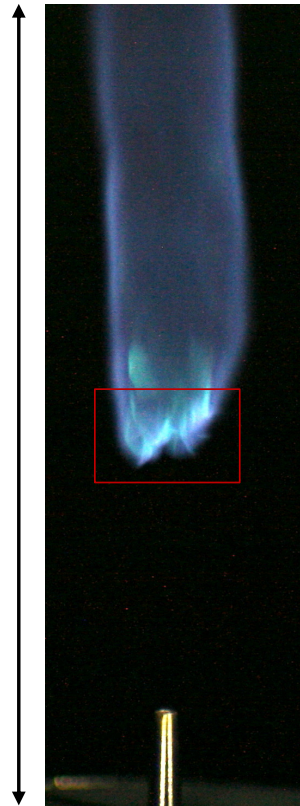
Pulse Burst + TPIV laser systems



High-Speed 3D LIF and Velocity at Base of Lifted Jet Flame

Lifted Dimethyl
Ether(DME)/Air
Jet Flame

102 mm



Summary on velocity measurements

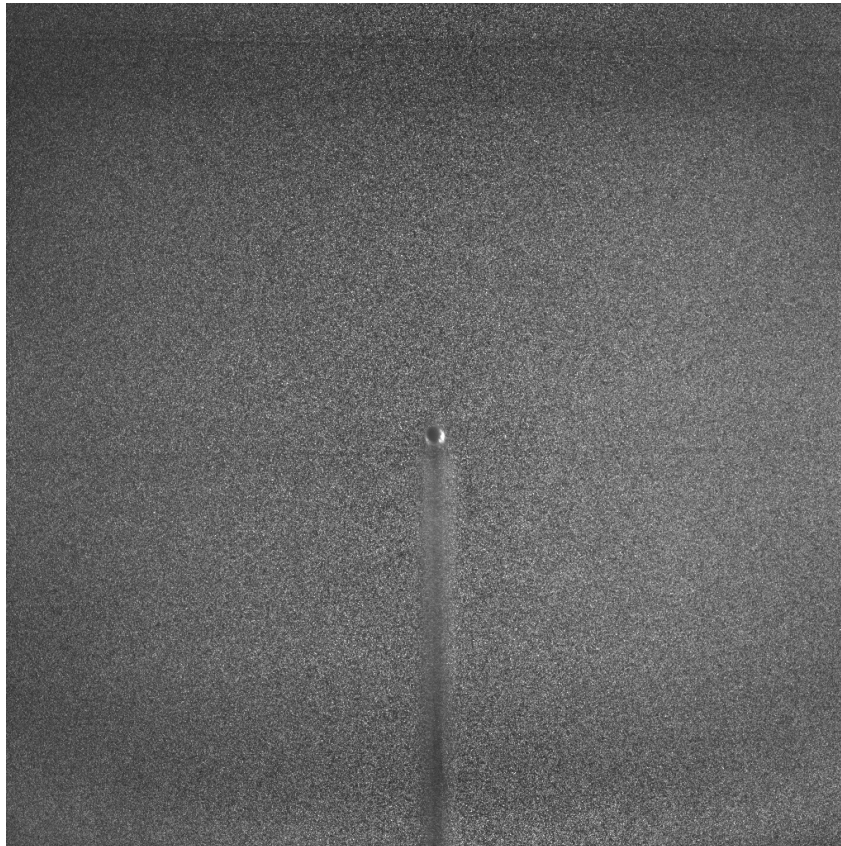
- LDA and PIV are complementary diagnostics for turbulent flames
- LDA extends to higher frequencies (validation data)
- PIV better as 2D images, combinations with scalar diagnostics, especially PLIF
 - Shake the Box and optical methods are variational methods for particle tracking/particle velocity beyond correlation methods
- Molecular methods (for places where particles can't go, e.g., high speed test facilities)
 - Narrowband Rayleigh; Doppler shift in scattered light changes throughput of molecular filter
 - Flow tagging; photodissociation in a grid of pulsed laser lines, followed by PLIF after ΔT (Miles et al., Pitz et al.)
 - Thermal grating velocimetry: crossed lasers beams excite seed molecule, creates thermal grating (Ewart et al.): only for very high velocities
 - FLEET: plasma generation and convection (Donehy et al.)
 - ...

Mie scatter for flame position and velocity: Measuring premixed laminar flame speeds

$$S_f = \frac{dR_f}{dt}$$

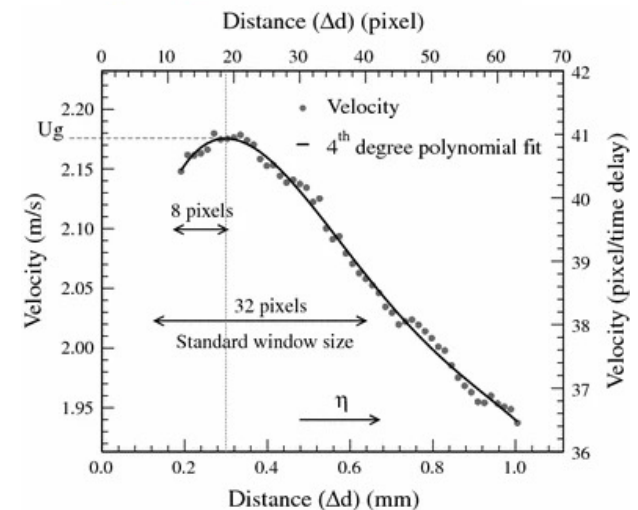
$$K = \frac{2}{R_f} \frac{dR_f}{dt}$$

CH₄-air
Φ = 1.0
P = 1 bar
T = 300 K



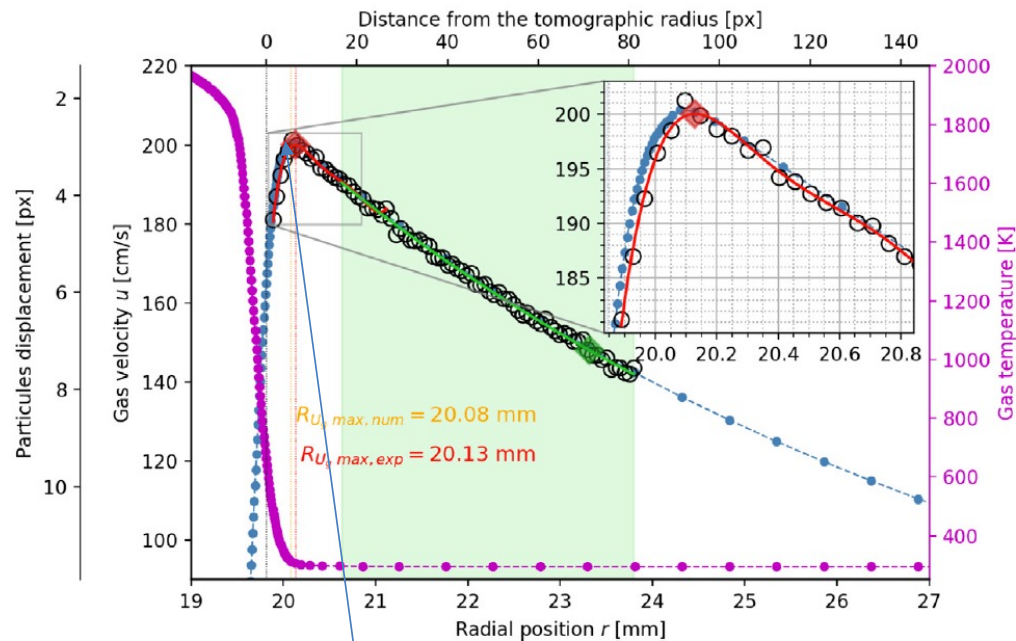
Courtesy: B. Renou,
INSA-Rouen

$$s_d = S_f - \underbrace{u_u}_{\text{Tricky to measure}}$$

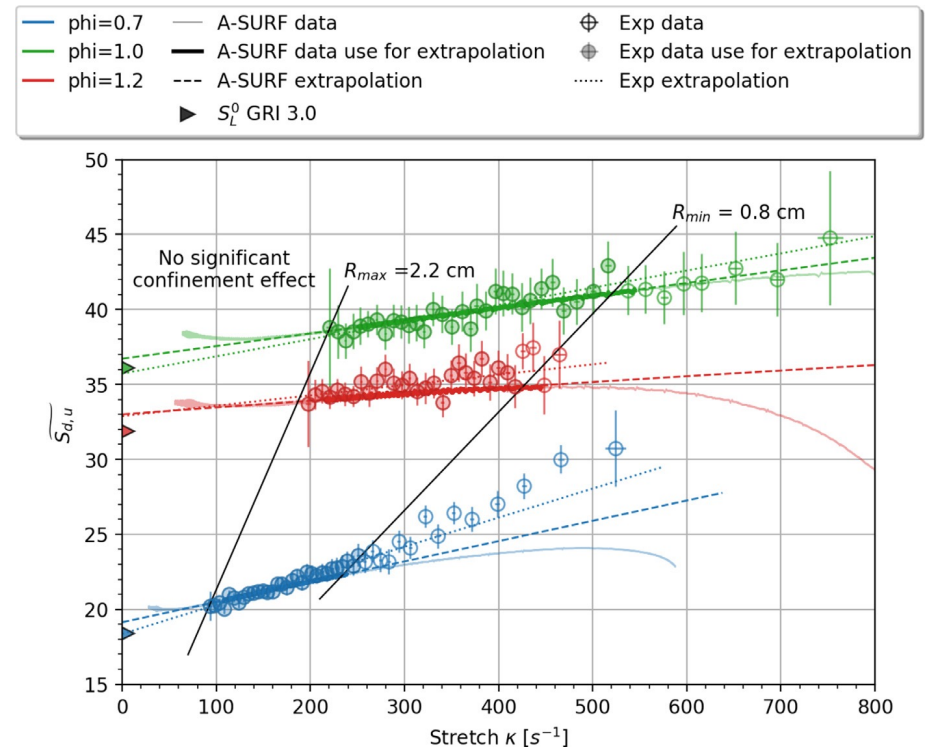


Balusamy, S., Cessou, A. & Lecordier, B.
Exp Fluids 50, 1109 (2011)

Direct measurements of displacement speed

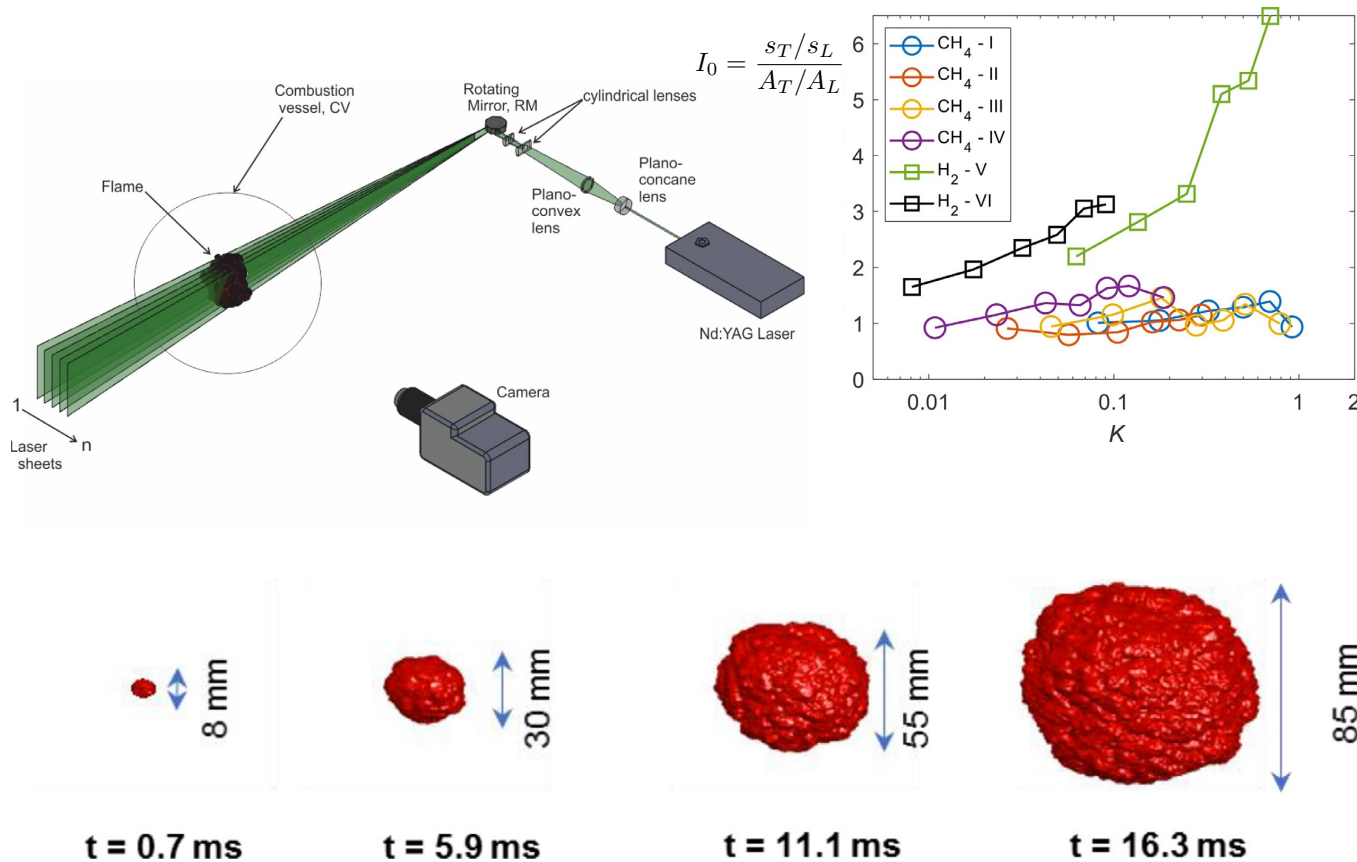


$$\tilde{s}_{d,u} = S_f - u_u$$

Methane/air flame ($\Phi = 1.0$)

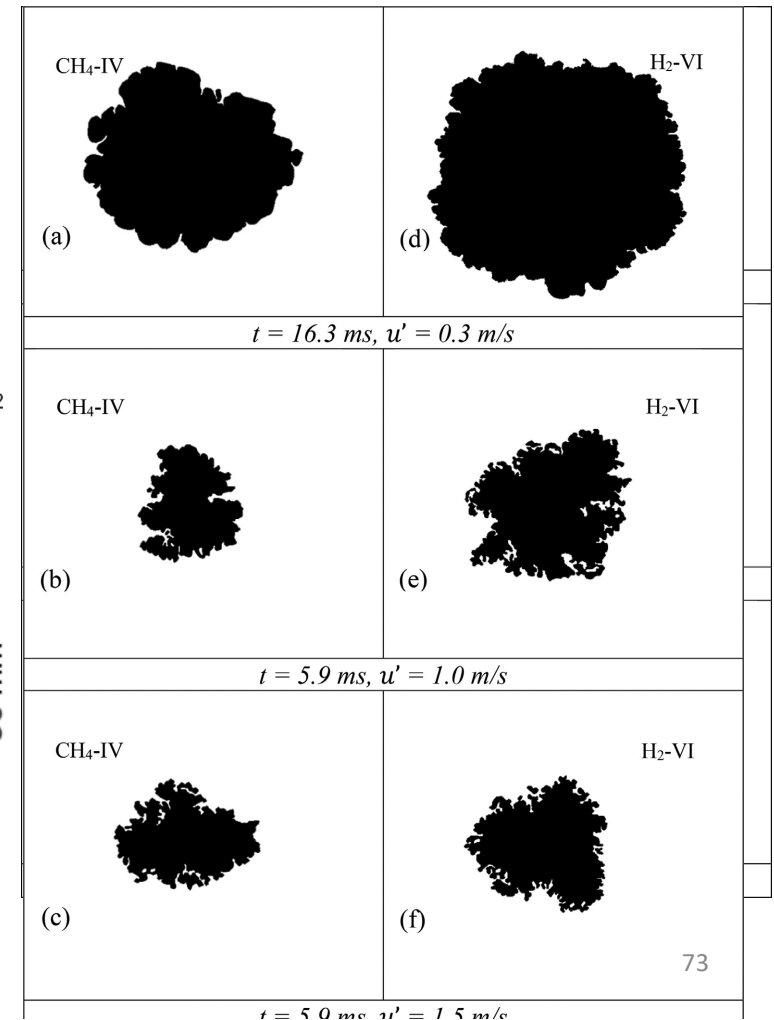
Clavel, M-E, Vandel A , Modica V, Chen Z , Varea, E, Moureau, V , Renou, B, Comb. Flame 235,, 111720 (2022).

3D Mie scatter for flame position and velocity

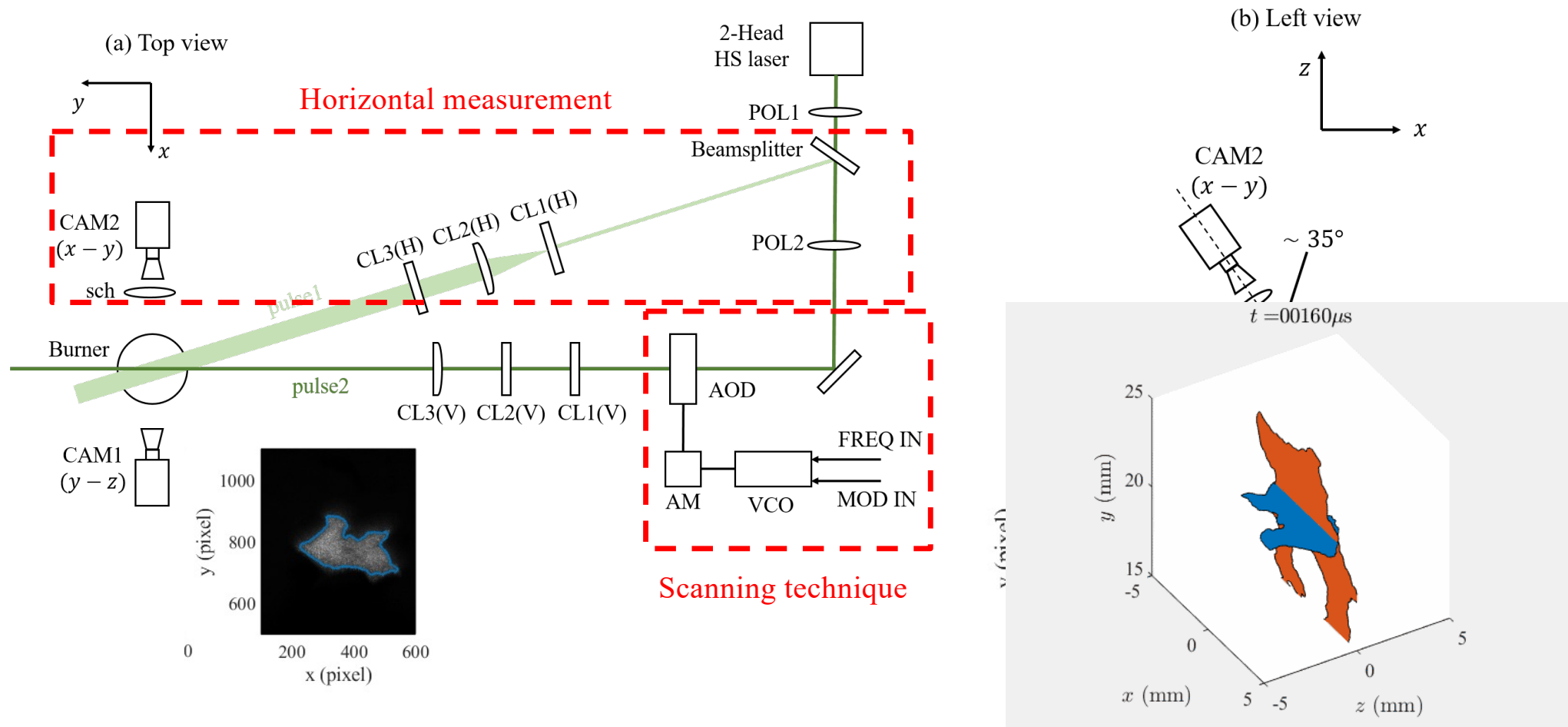


[Ahmed et al., Comb. Flame 233, 111586 \(2021\)](#)

Brazil Winter School 2025

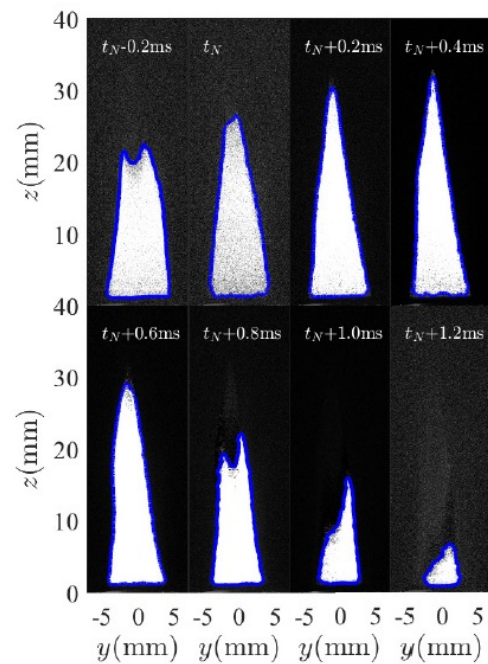


Laser-based 3D reconstruction of flame surfaces

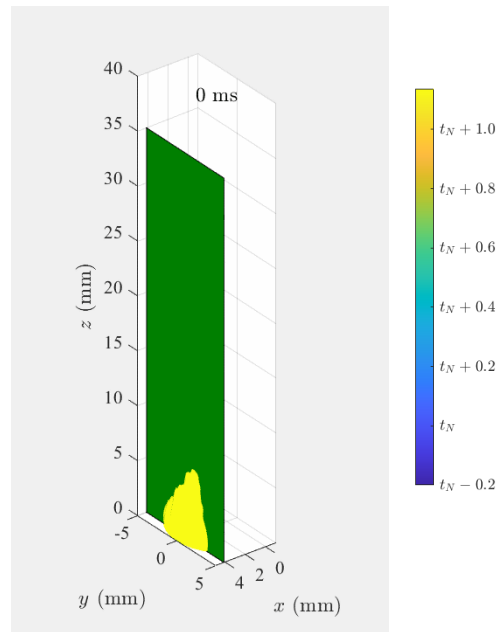


Laser-based 3D reconstruction of flame surfaces

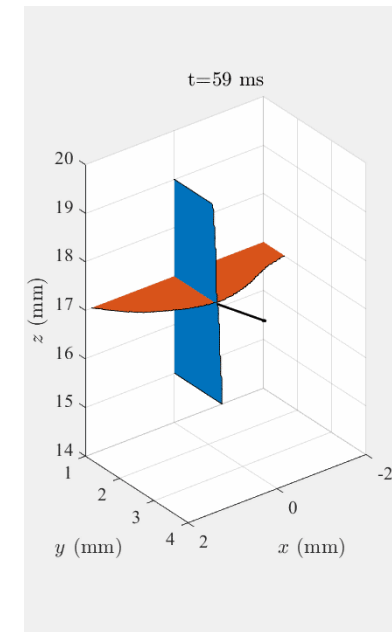
Individual scanned images



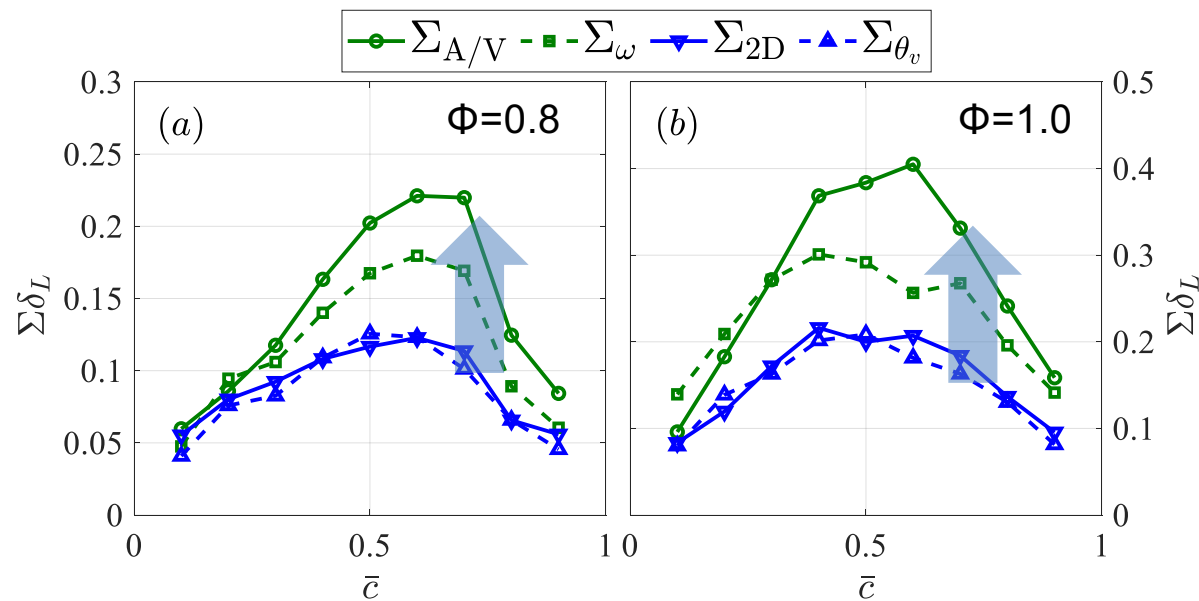
Surface reconstruction



3D normal



Comparison of local flame surface density via two techniques

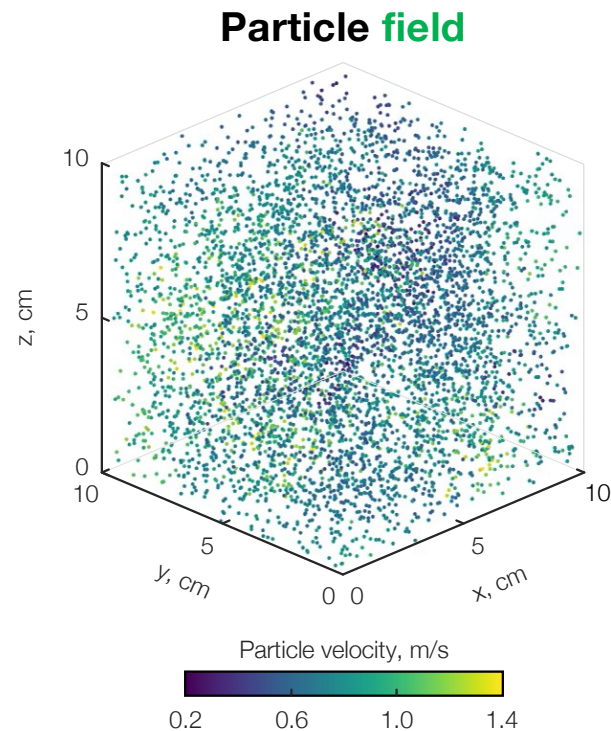


Local FSD measured via two techniques, scanning technique and cross-planar technique along the intersection line, compared with 2D FSD measured from central plane

[Zheng, Y., Weller, L. and Hochgreb, S., 2023. 3D Flame surface density measurements via orthogonal cross-planar mie scattering in a low-turbulence bunsen flame. \(2023\), Proc. Comb. Inst. 39: 2369-2377](#)

Zheng, Y. et al., 3D flame surface measurements in turbulent Bunsen flames via scanning and orthogonal cross-planar technique (2023), Comb. Flame, submitted.

Lagrangian particle tracking



1. Seeding, illumination & imaging

Tracer particles are seeded into the flow (or arise spontaneously); the particles are illuminated with a light source and then imaged

2. Localization & tracking

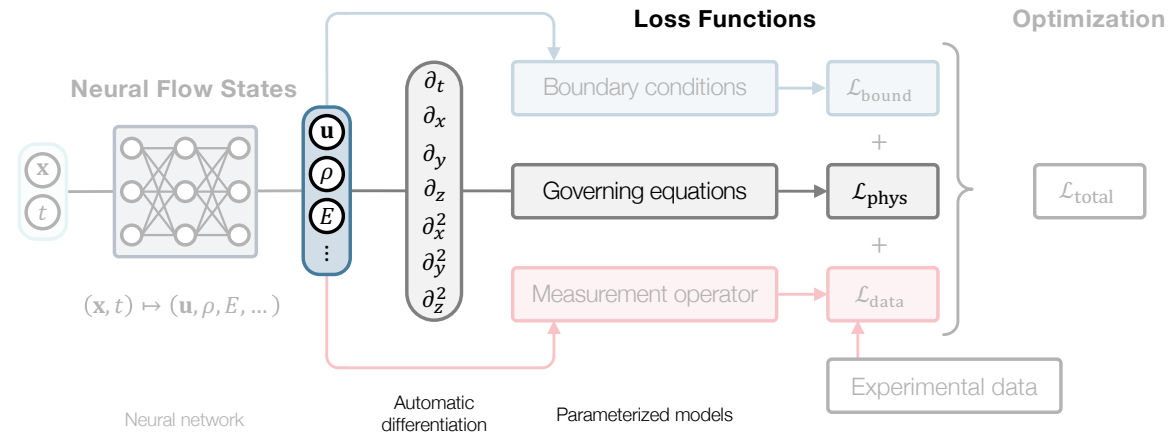
Each particle is localized in 3D space by triangulation or numerical refocusing, after which the particles are linked across frames to produce Lagrangian tracks

3. Flow reconstruction & analysis

Eulerian flow states, comprising velocity, pressure, and other fields, are reconstructed from the tracks, and this information may be analyzed to study the underlying flow physics

Courtesy: S Grauer

How We Do It: ‘Neural Data Assimilation’



Governing equations

- *Suitable governing equations* are evaluated, and the residuals are added up to form a “*physics loss*”

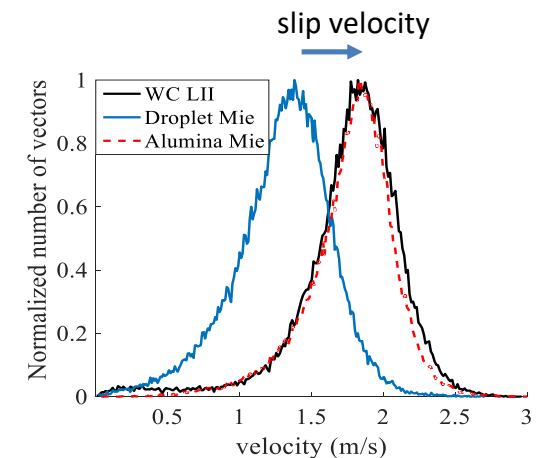
$$\mathcal{L}_{\text{phys}} = \frac{\dim(\mathbf{e})^{-1}}{|\mathcal{V} \times \mathcal{T}|} \int_{\mathcal{T}} \iiint_{\mathcal{V}} \|\mathbf{e}(\mathbf{x}, t; \boldsymbol{\theta})\|_2^2 d\mathbf{x} dt$$

$$\begin{aligned} e_c &= \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) \\ \mathbf{e}_m &= \frac{\partial (\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \mathbf{u}^T) + \nabla p - \nabla \cdot \left[\mu (\nabla \mathbf{u} + \nabla \mathbf{u}^T) - \frac{2}{3} \mu (\nabla \cdot \mathbf{u}) \mathbf{I} \right] \\ e_e &= \frac{\partial (\rho E)}{\partial t} + \nabla \cdot [(\rho E + p) \mathbf{u}] - \nabla \cdot (\kappa \nabla T) - \nabla \cdot \left\{ \left[\mu (\nabla \mathbf{u} + \nabla \mathbf{u}^T) - \frac{2}{3} \mu (\nabla \cdot \mathbf{u}) \mathbf{I} \right] \cdot \mathbf{u} \right\} \\ &\vdots \end{aligned}$$

Courtesy: S Grauer

New developments in particle scattering methods

- Use of phosphorescent particles for PIV (get temperature and velocity simultaneously) (see F. Beyrau, B. Fond, L. Fan's papers)
- Use laser induced incandescence of carbide particles (energies are enough to light them up in the gas phase, not in liquid phase, so get separate velocities of gas and liquid phase)
- 3D/Lagrangian reconstruction of particle velocities for higher resolution (computationally expensive, but effective)
- Fast velocity reckoning using optical methods via a reference image



[Fan, L. et al., Proc. Comb. Inst. 38:1\(2021\),](#)

Questions



Why do I need to seed the flow?



How does autocorrelation work?



What is the limit of domain size for PIV?



Why should I care about velocities?



Does it matter what size the particles are?



What particles should I use?

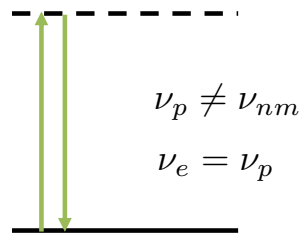
Pause



Outline

- Why and how we measure
- Fundamentals of optical diagnostics
 - Light sources and signal connection
 - Detection methods
 - Light-molecule interactions
- Incoherent
 - Scattering and particle velocimetry
 - Rayleigh scattering
 - Raman scattering
 - Absorption
 - Laser induced fluorescence
 - Laser-induced incandescence
- Coherent techniques
 - CARS
 - LIGS

Rayleigh scattering: molar density measurements



Elastic scatter:

Rayleigh

(non-resonant
induced dipole)

$$\frac{I}{I_0} = \frac{\sigma}{\sigma_0} \frac{N}{N_0} = \frac{\sigma}{\sigma_0} \frac{T_0}{T}$$

signal

$$I_s = \eta \sigma I_i V N$$

collection efficiency

Incident

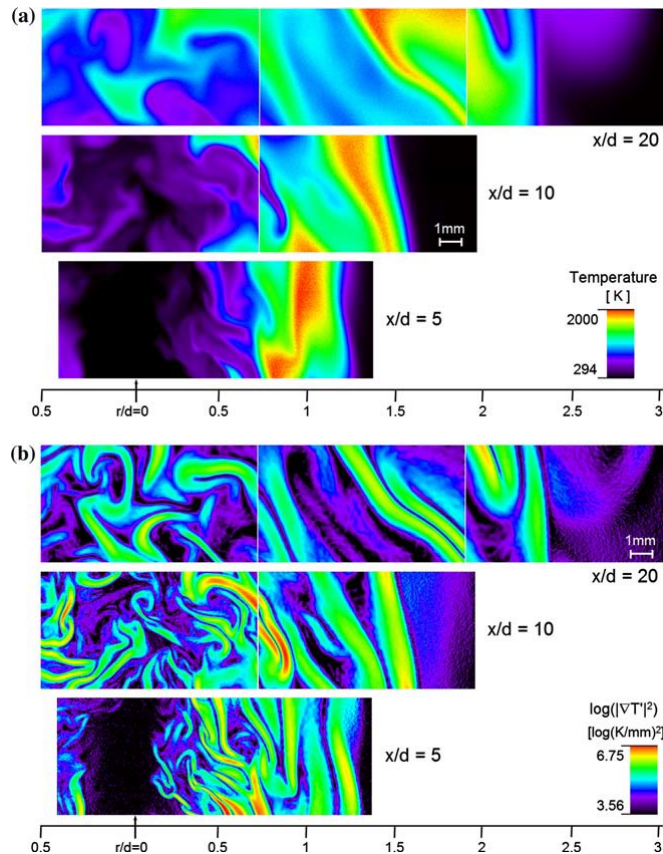
Probe volume

$$N = \frac{p}{kT}$$

- Mean scattering cross section of the mixture
- Weakly dependent on temperature and composition
- Proportional to $(a\nu)^4$, where a is molecular diameter: but optical collection efficiency often lower at high ν

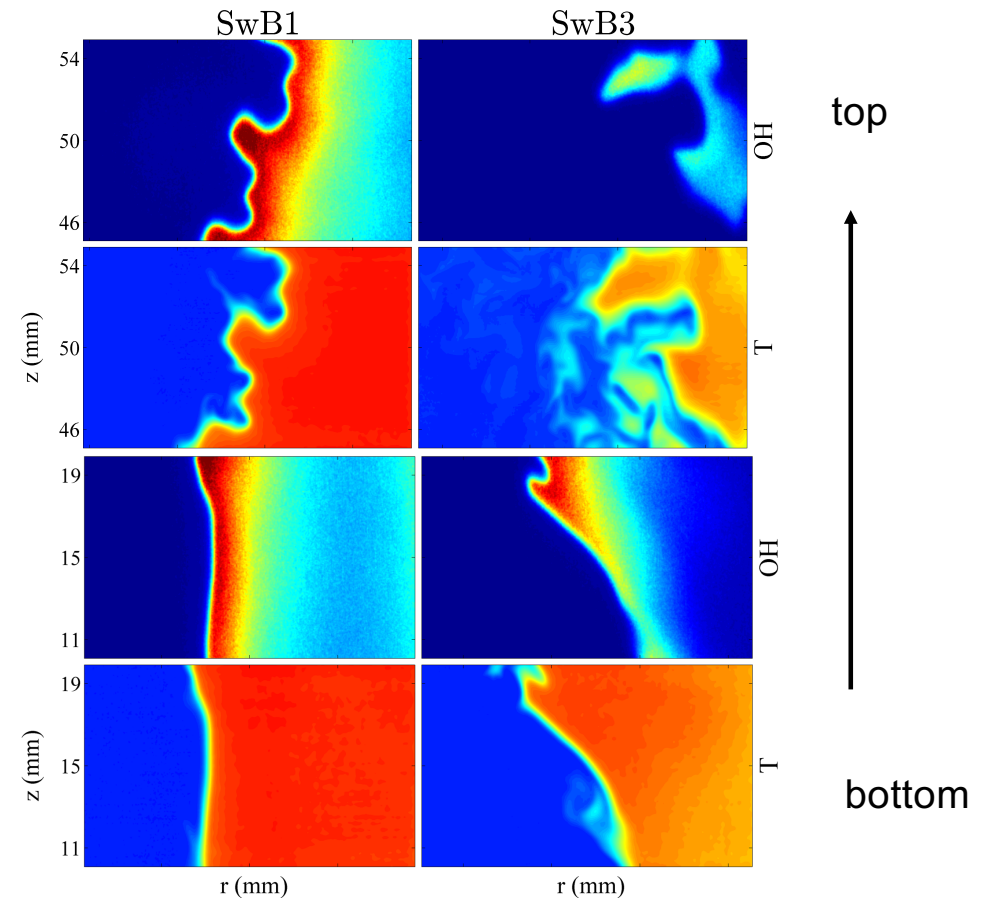
- Sensitive to elastic scattering from surfaces and particles:
- Not usually suitable for enclosures
- Gated ICCD camera required for luminous flames
- Relatively easy to set up
- Weak signal (requires ~ 1 J/pulse for imaging 2x2 cm area)

Temperature via Rayleigh scattering



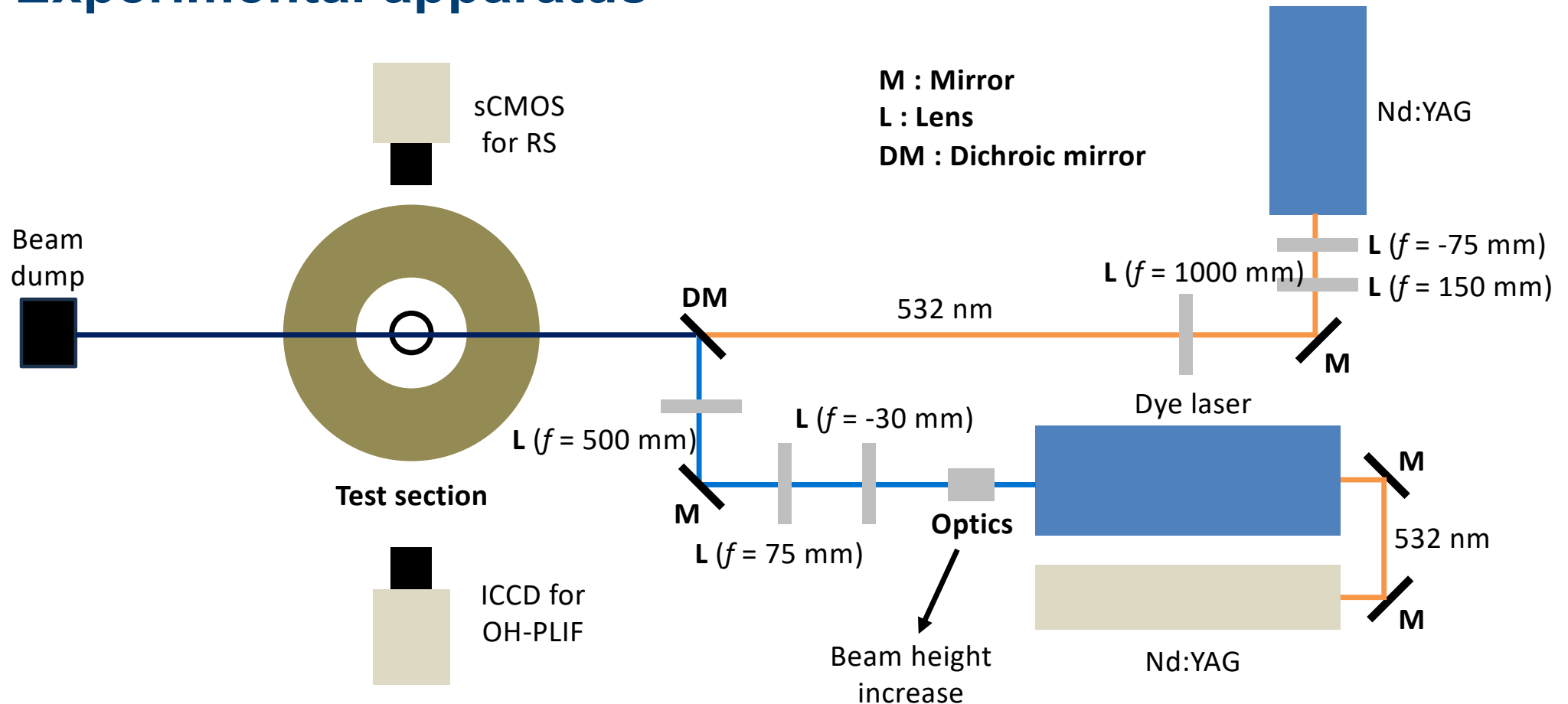
Single-shot measurements of temperature (a) and the squared gradient of the temperature fluctuation $|\nabla T'|^2$ (b), at the downstream locations.

Frank, J.H., Kaiser, S., Exp. Fluids. 44 (2007) 221–233



Kamal, M. M., Coriton, B. et al., Proc. Comb. Inst. 36 (2017) 1957–1965

Experimental apparatus



Filters for Rayleigh scattering (Methane)

$$S_{R,N}^S = \frac{I_F(x, y, t) - (\overline{I_{FB}}(x, y) + \overline{I_S})}{\overline{I_A}(x, y) - (\overline{I_D}(x, y) + \overline{I_S})} \quad SNR = \frac{\overline{S_{R,N}^S}}{\sigma(S_{R,N}^S)}$$

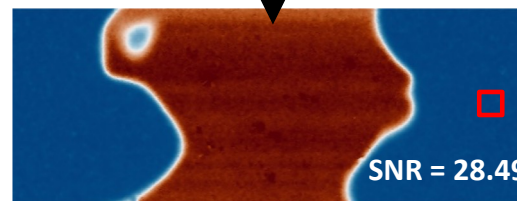
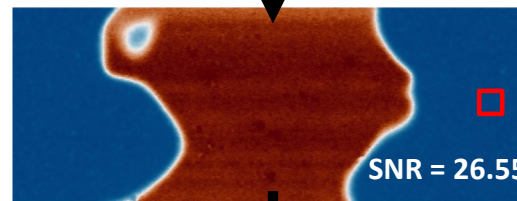
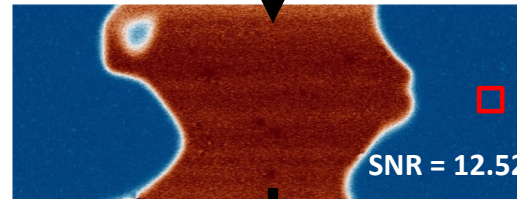
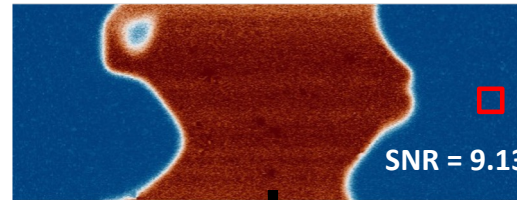
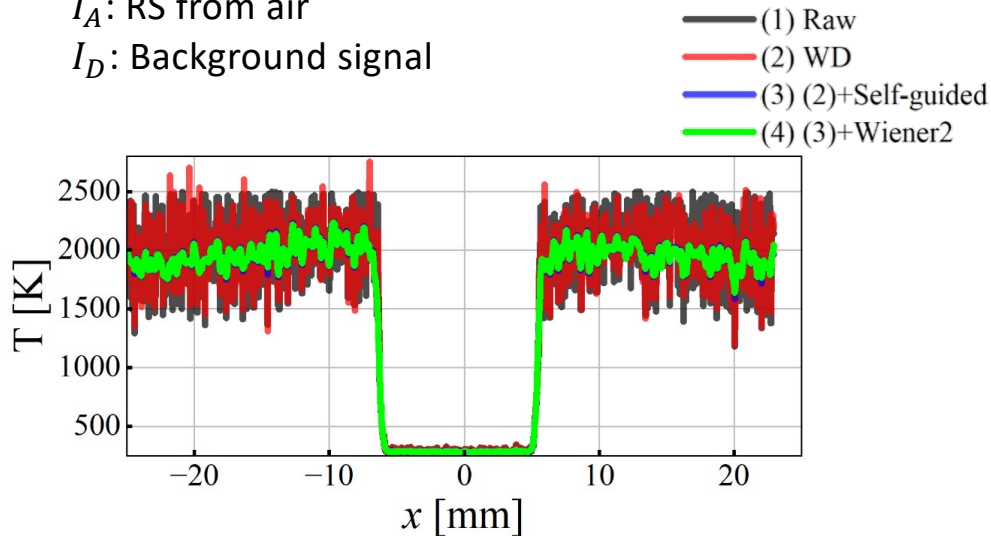
I_F : RS from flame

I_{FB} : Flame luminosity

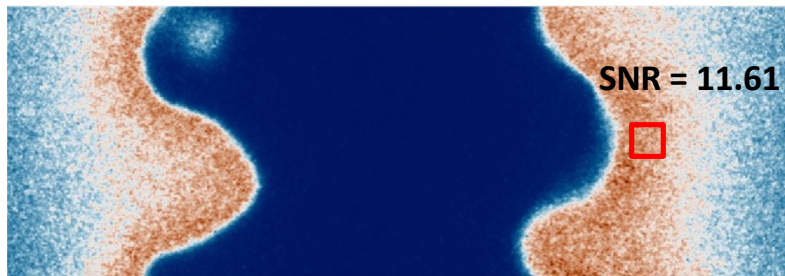
I_S : Un-rejected spurious scattering

I_A : RS from air

I_D : Background signal



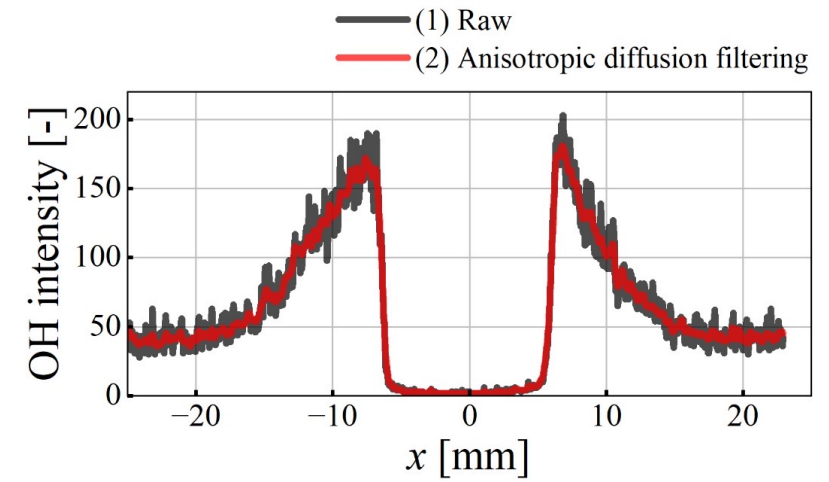
Filters for OH PLIF



1. Raw OH PLIF

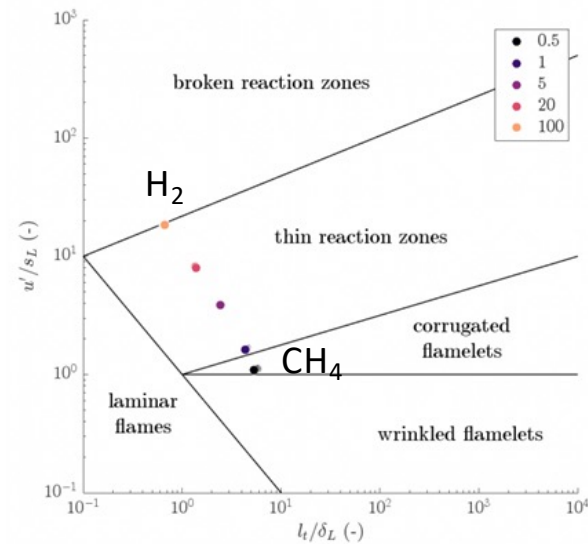
2. Anisotropic diffusion filtering

Number of iterations = 20

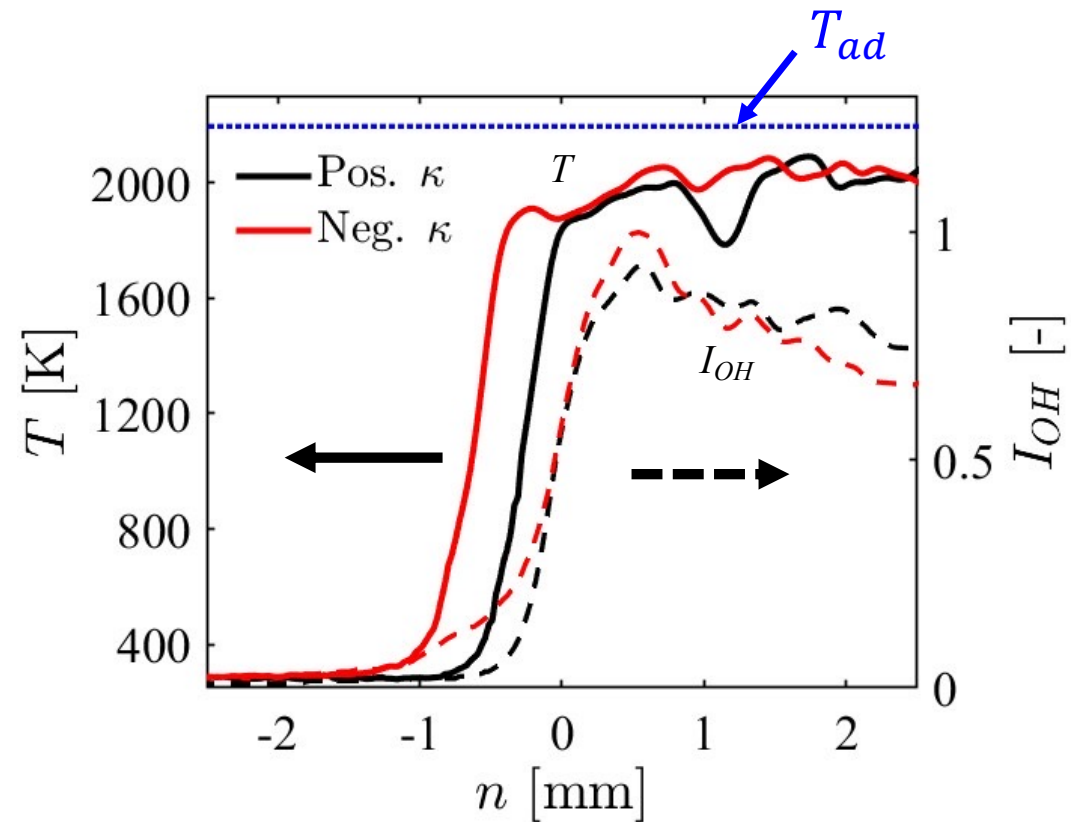
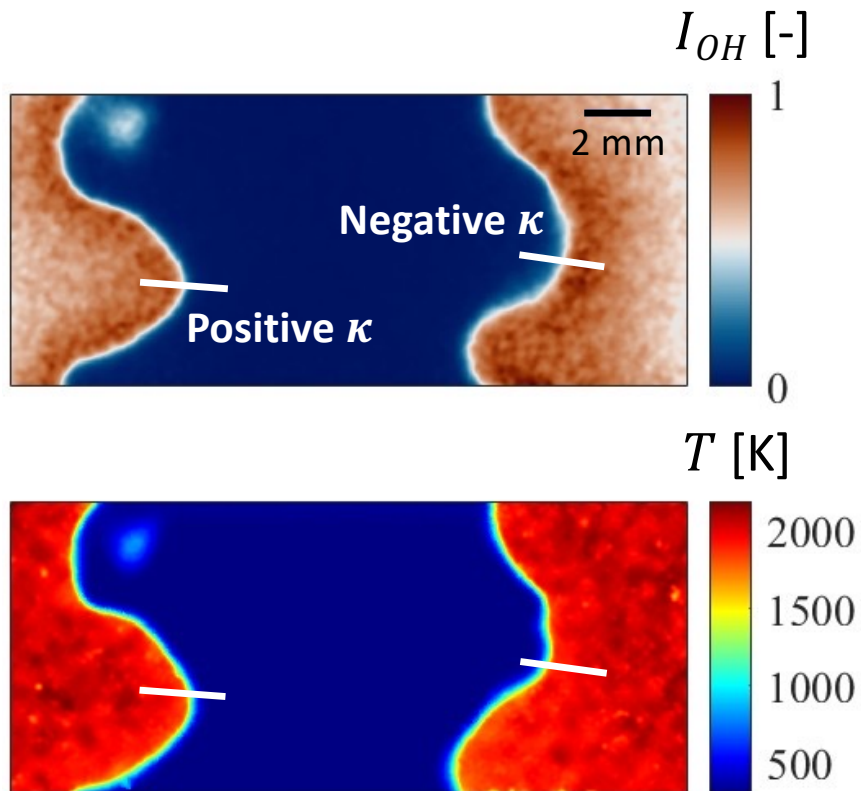


Test condition (RS + OH-PLIF)

Test number	Ka	Main flame					Pilot flame			
		Air [slpm]	CH ₄ [slpm]	H ₂ [slpm]	H ₂ %	T_{ad} [K]	Air [slpm]	CH ₄ [slpm]	Eq.ratio	T_{ad} [K]
Test 1	0.5	35.97	3.61	0	0	2194	143.8	12	0.79	1988.5
Test 2	100	34.98	0	4.62	100	1224.5	143.8	12	0.79	1988.5

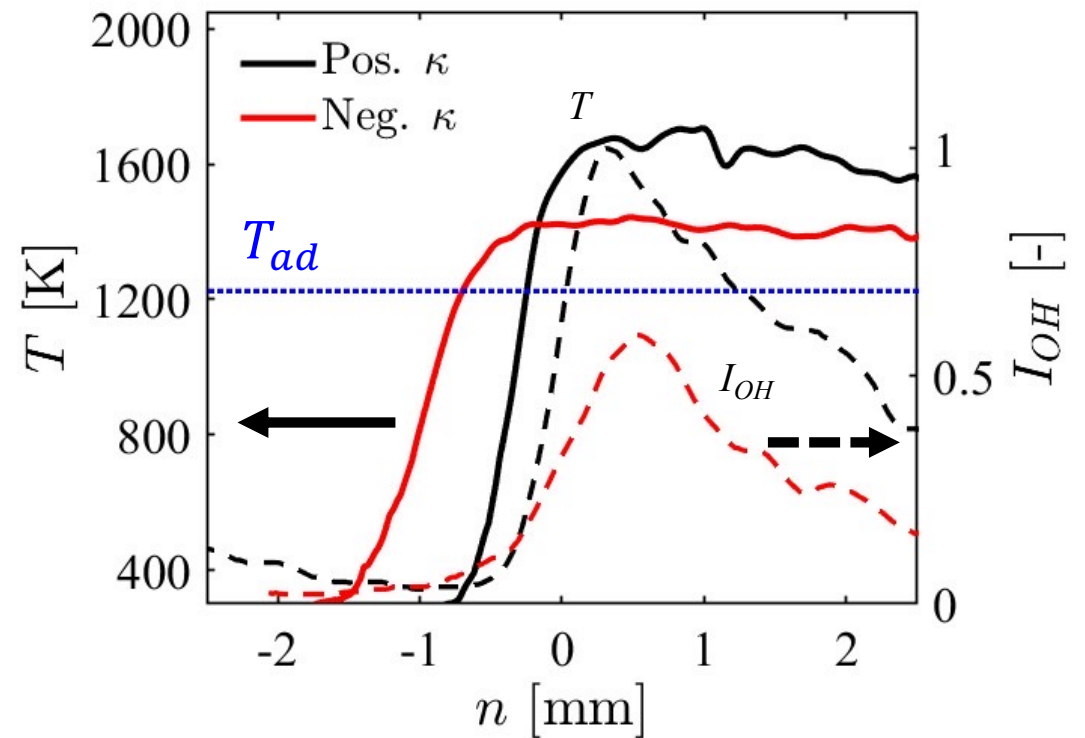
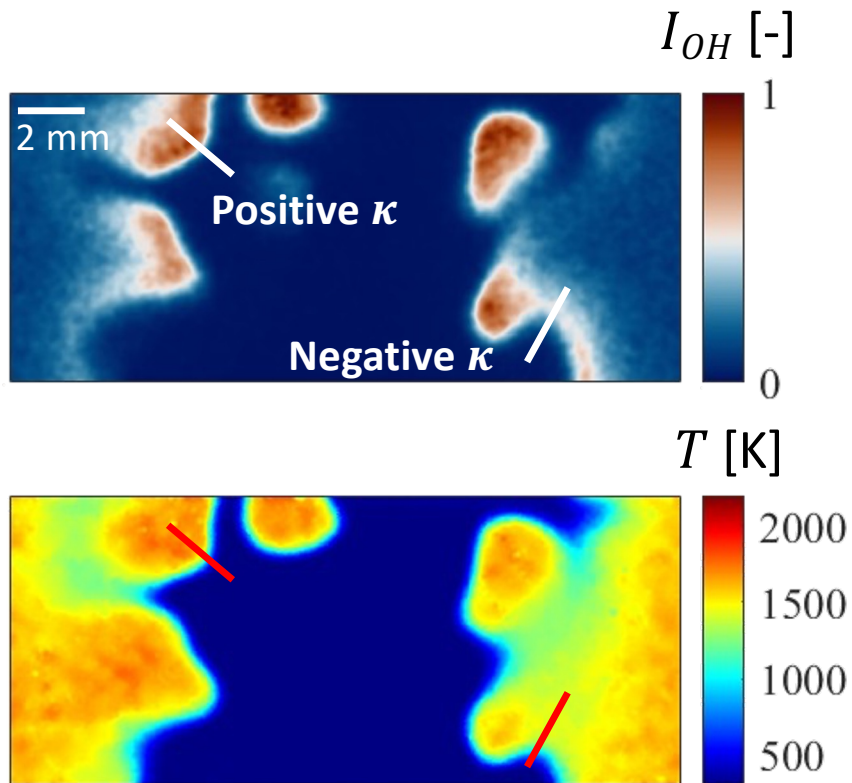


Flame structure : methane



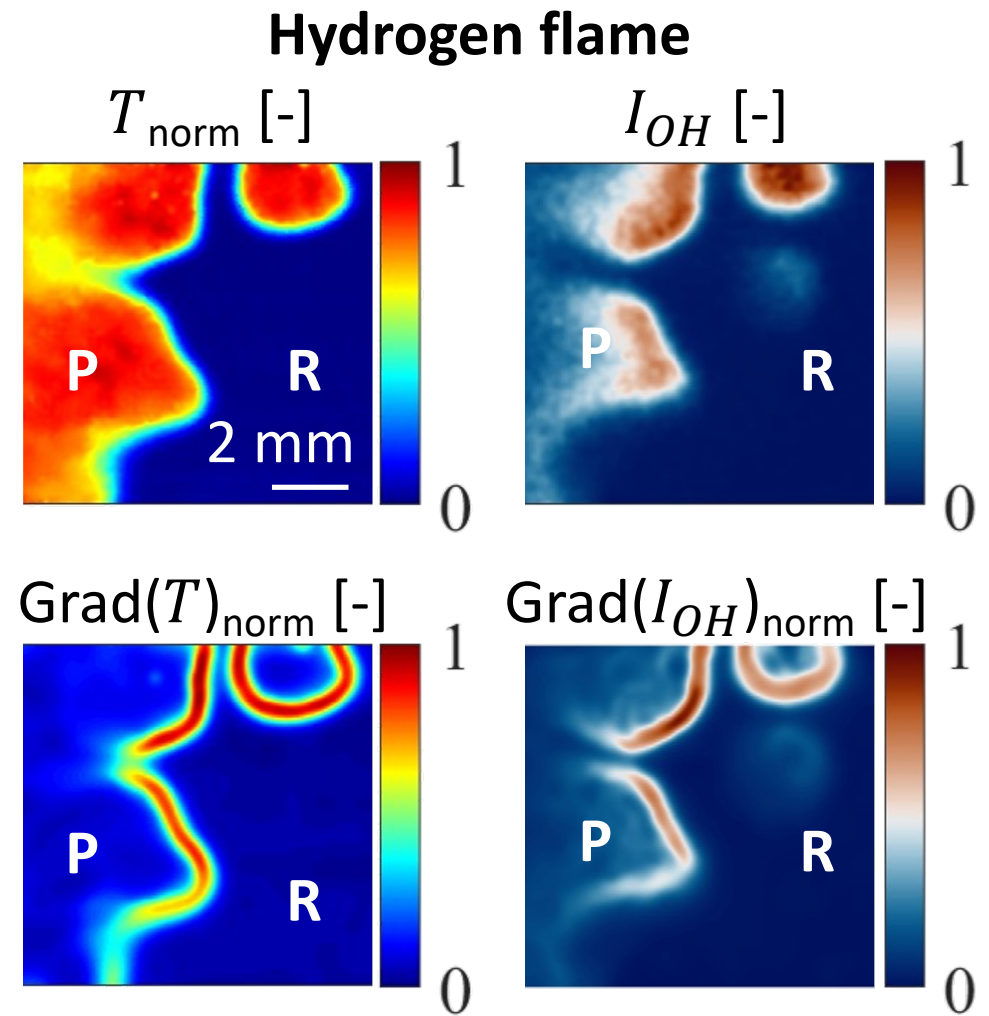
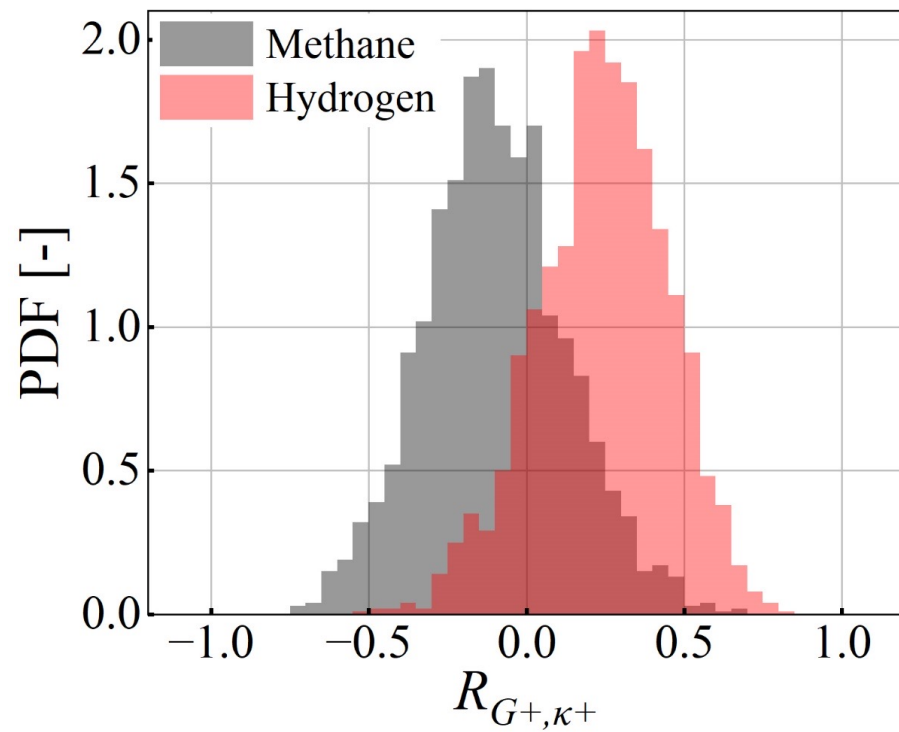
Grad(T) & Grad(OH) are similar regardless the curvature.

Flame structure : hydrogen

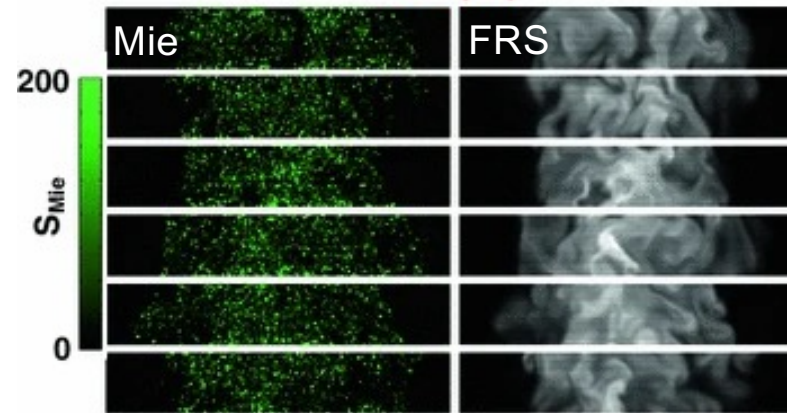
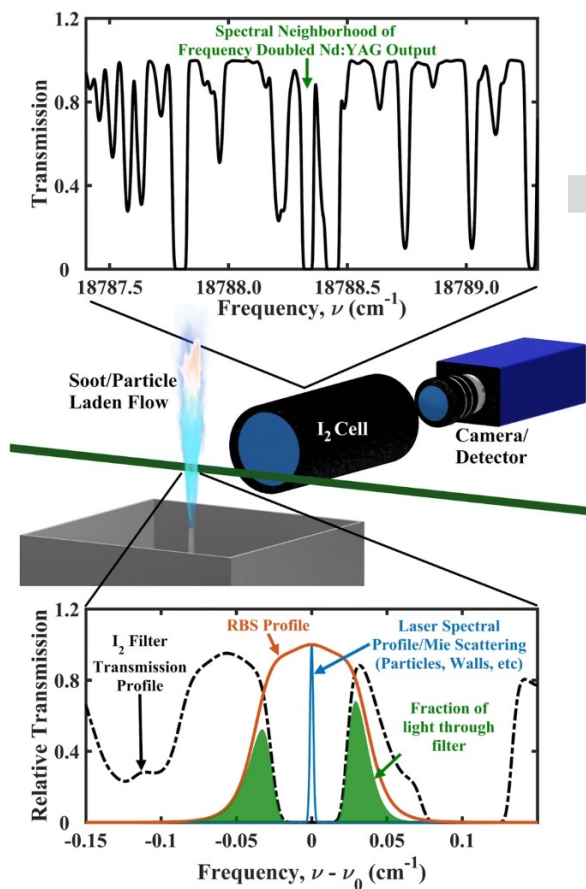


Grad(T) & Grad(OH) decrease for negative curvature.

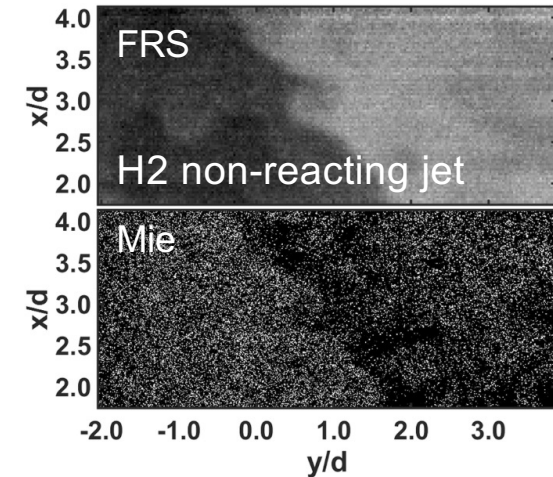
Curvature and gradient



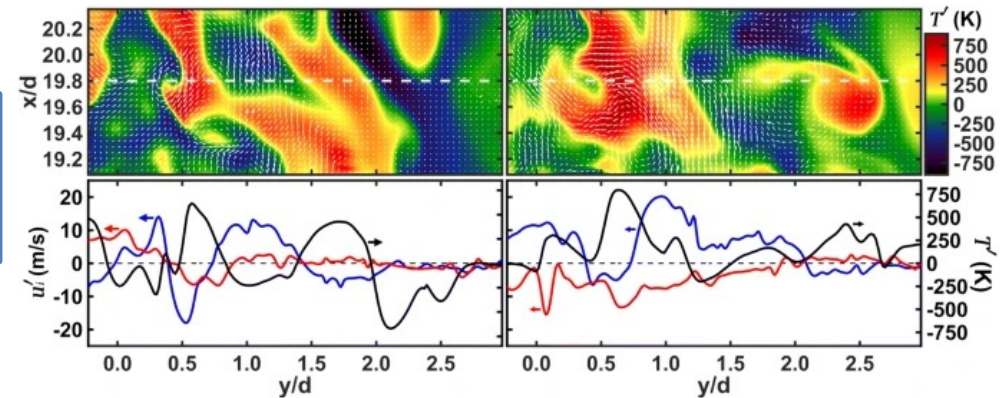
Filtered Rayleigh scattering: separating Mie background



$(F_{Vp} \sim 5 \text{ ppm}) \text{C}_3\text{H}_8$ jet



Simultaneous
temperature and
velocity measurements



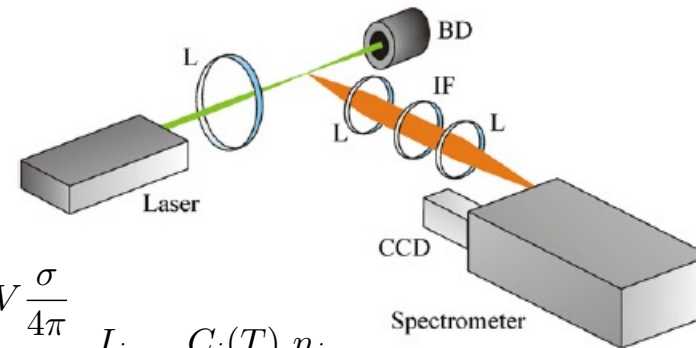
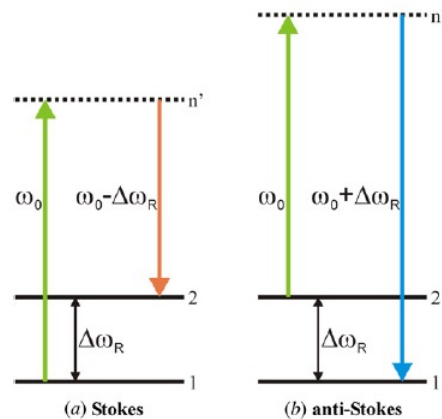
[Sutton, J.A., Patton, R.A. *Appl. Phys. B* **116**, 681–698 \(2014\).](#)
[Trueba-Monje & Sutton, *AIAA Sci Tech. Forum* 2018](#)

[McManus, T.A., Sutton, J.A *Exp Fluids* **61**, 134 \(2020\).](#)

Outline

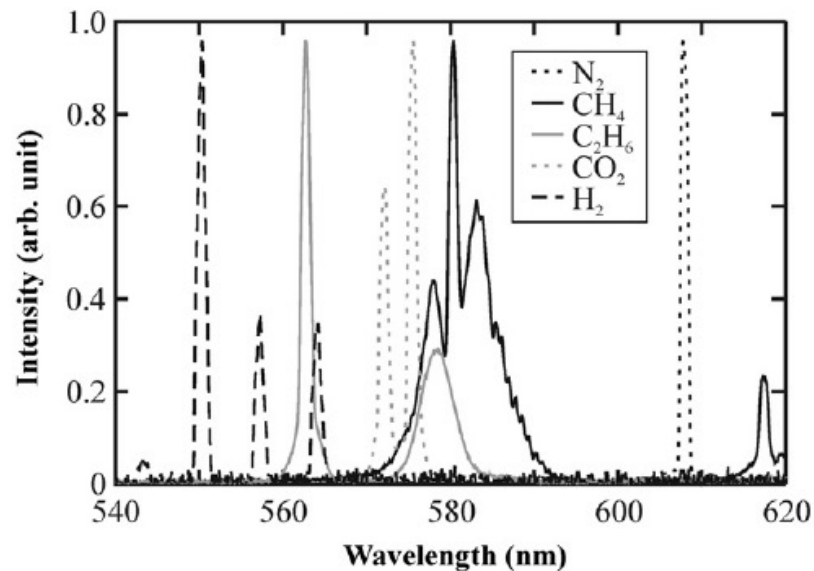
- Why and how we measure
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 - Raman scattering
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Species measurement: Raman



$$I_i = K n_i I_o V \frac{\sigma}{4\pi}$$

$$\frac{I_j}{I_i} = \frac{C_i(T)}{C_j(T)} \frac{n_i}{n_j}$$

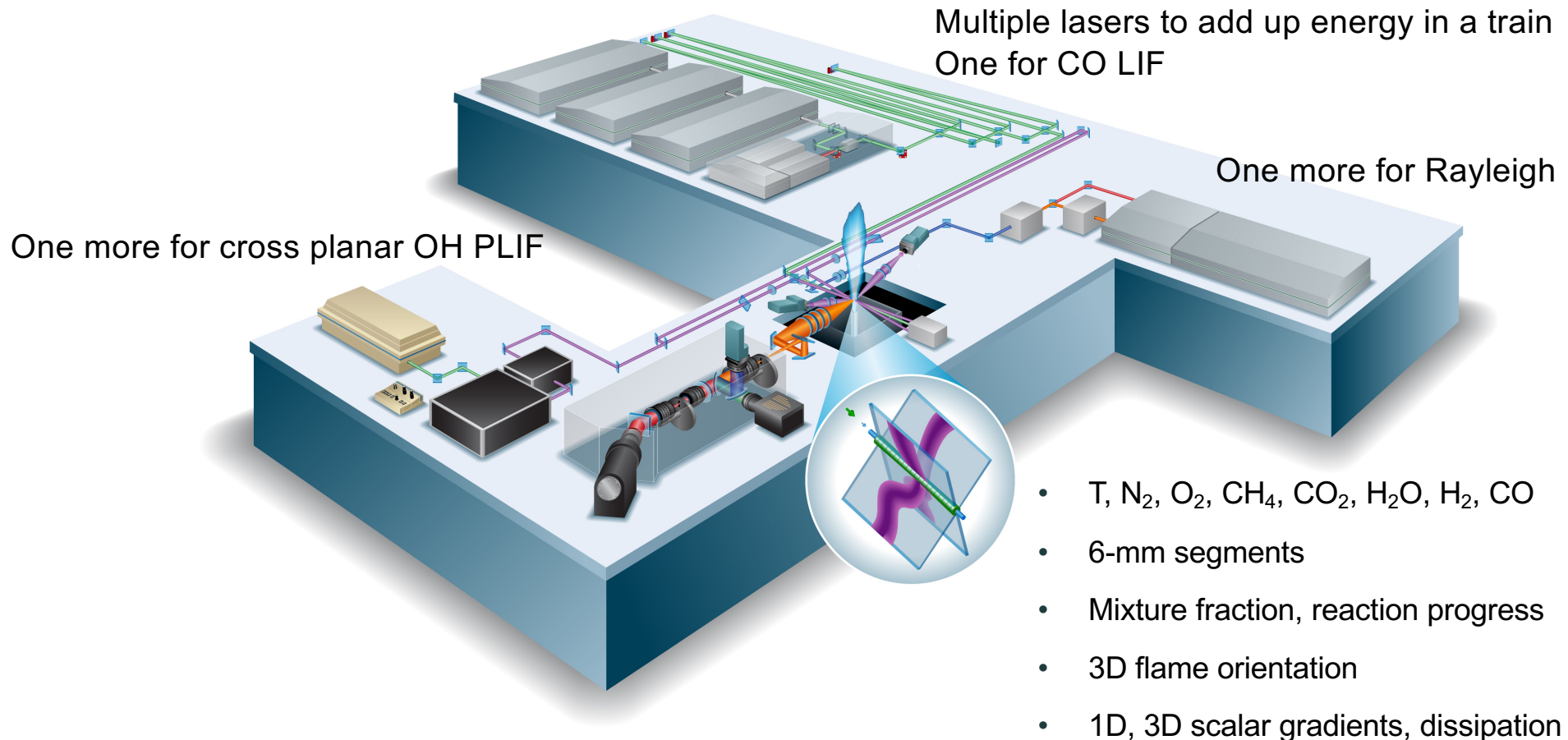


Many lines: all species simultaneously.
Line overlap!
Requires experience in selection of lines

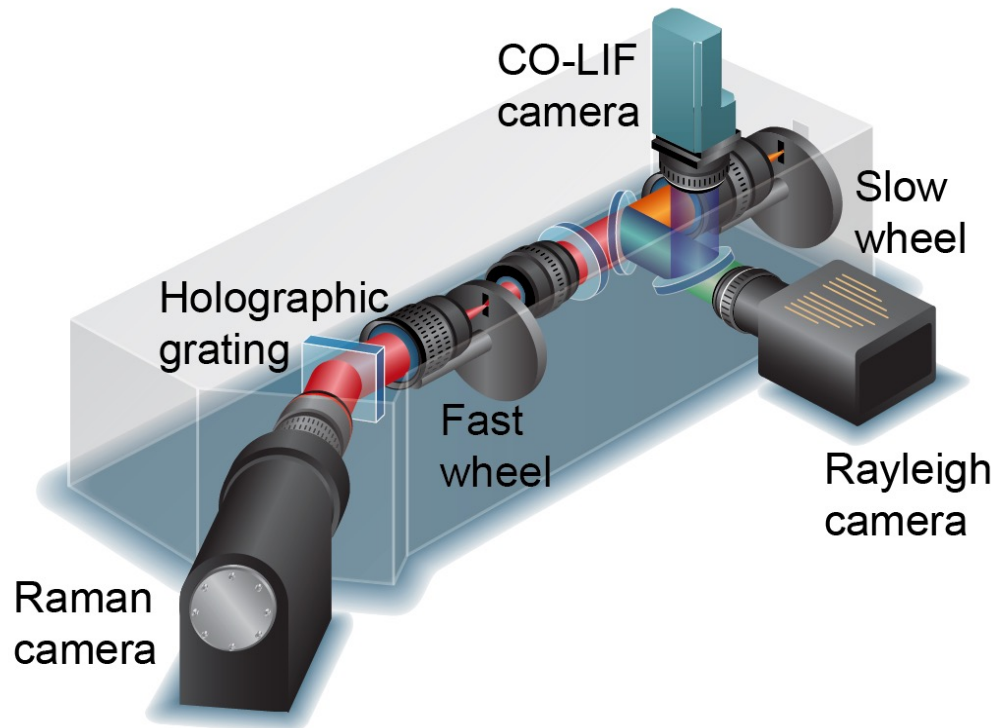
Very low overall yield (10^4)
Requires very high energies/pulse to remove
interferences in combustion (1-2 J/pulse)

Kiefer et al, Meas. Sci. Tech. (2008)

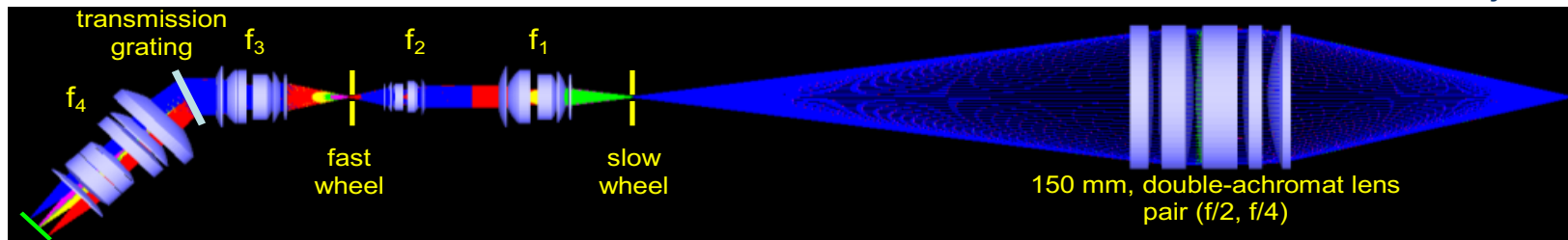
Sandia Raman-Rayleigh-CO LIF facility (R.I.P.)



Raman/Rayleigh/CO-LIF Detection System



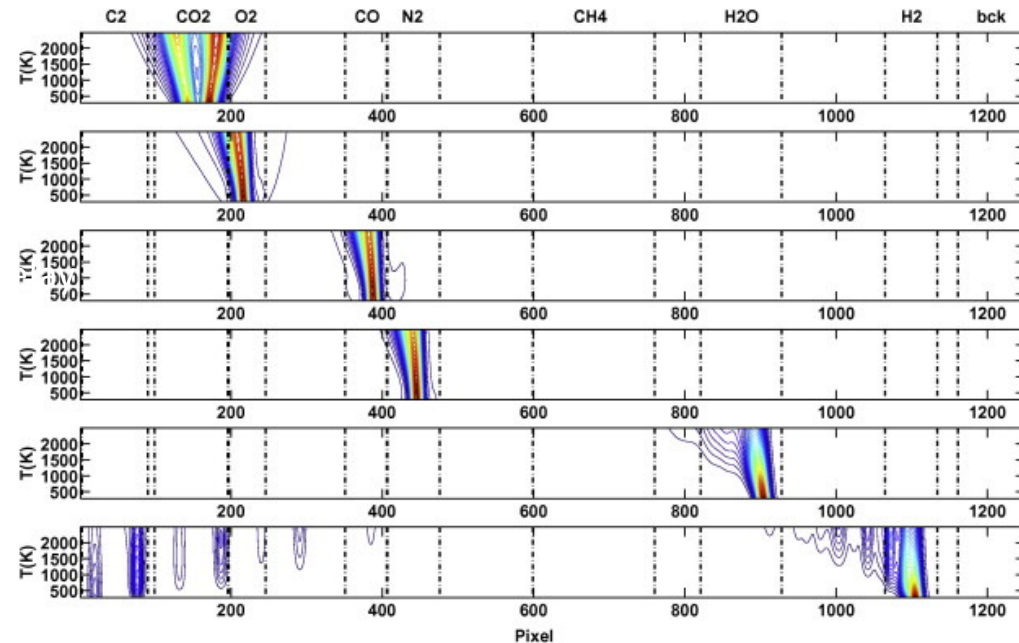
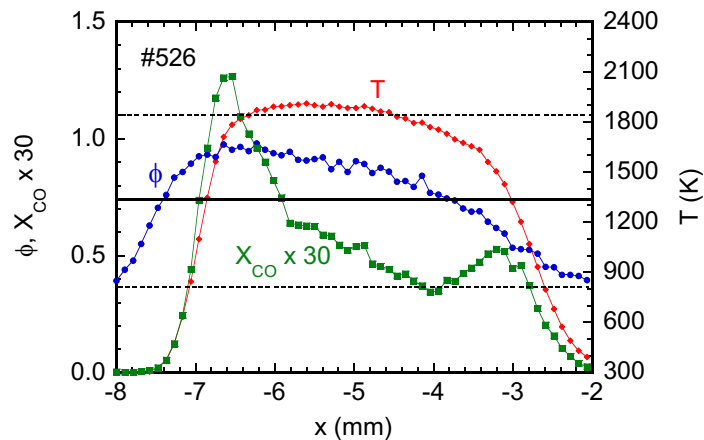
- Lenses (Raman system):
 - 85mm f/1.8, Nikon
 - 50mm f/1.4, “
 - 85mm f/1.4, “
 - 135mm f/2, Canon
- Custom grating (Kaiser Optical)
 - 1200 lines/mm
 - 90% efficiency at 625-nm
 - 76mm x 76mm
- Fast wheel:
 - 21000 rpm (3.9 μ s gate FWHM)
 - 150 W thermal load
- Slow wheel: 3000 rpm (300 μ s [shutter])
- PLL motor control +/- 400 ns jitter



Light collection and calibration

Fuest et al, PCI 33 (2011)

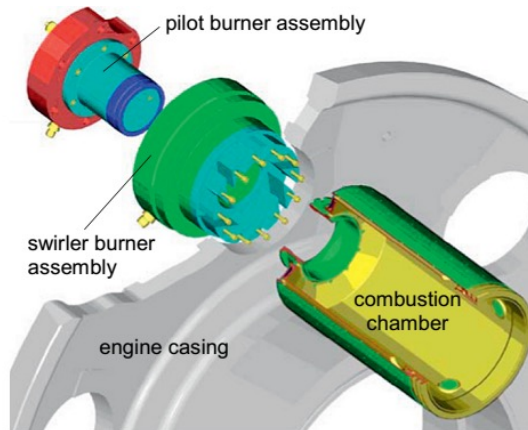
- Complex calibration depends on temperature, wavelength and experimental details.
- Needs sufficient experience for good quantitative results.



Modeled temperature response

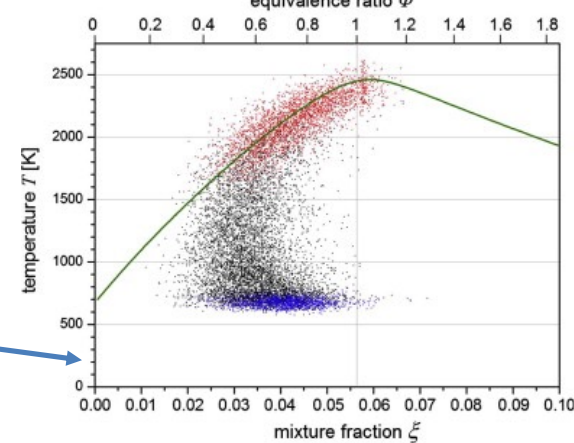
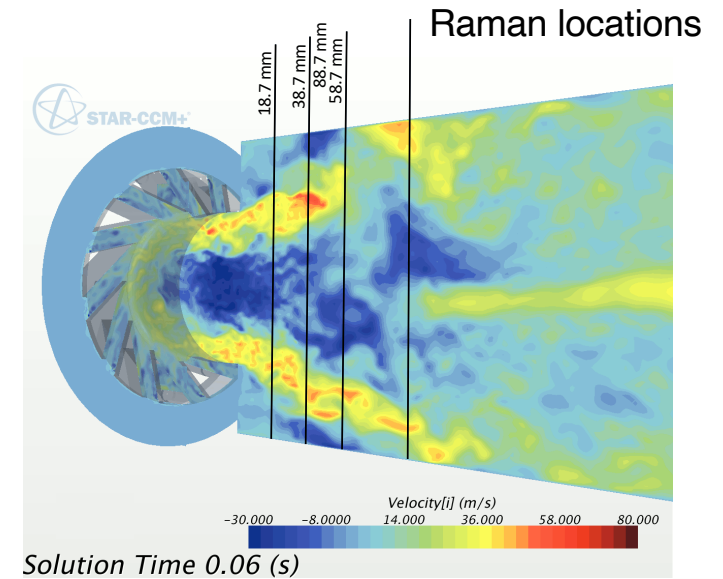
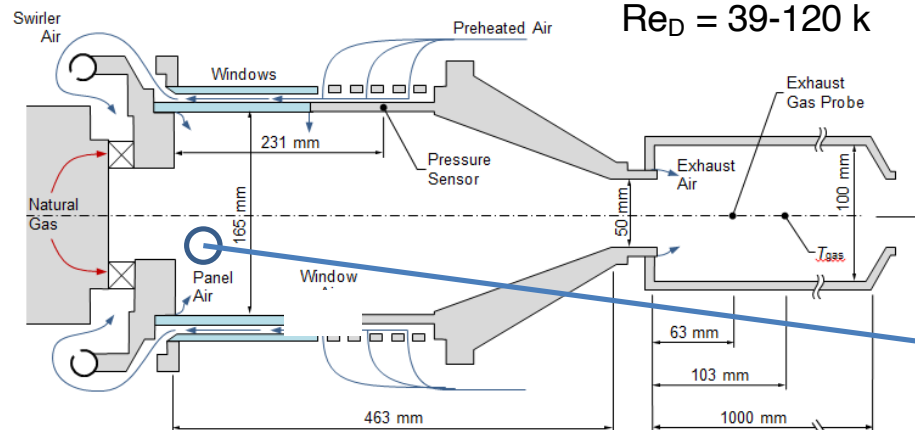
Single shot multi-species at high resolution

Industrial combustors: SGT-100 DLE



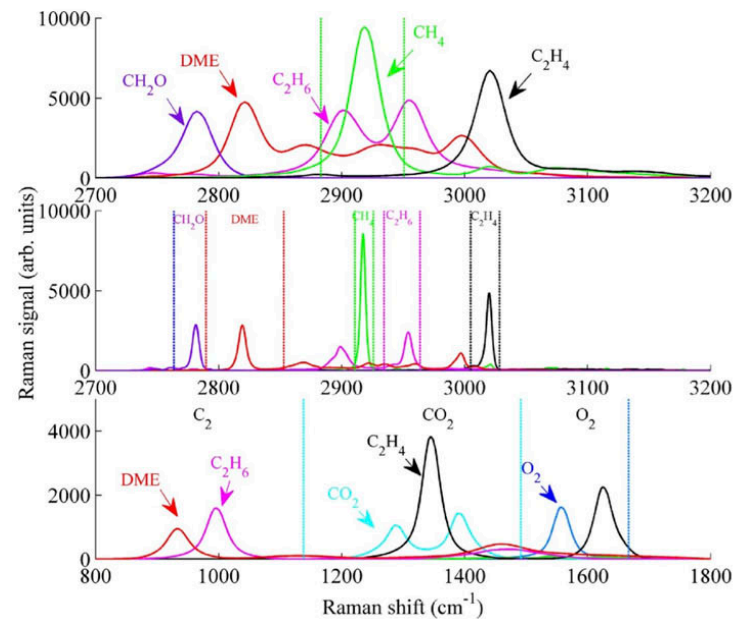
U. Stopper et al.
Comb. Flame 160 2103-2118 (2013)
U. Stopper et al.
Exp. Therm. Fluid Sci. 34 396 - 403 (2010)
U. Stopper et al. J. Eng. Gas Turbines
Power 131 021504 (2008)

$P = 3\text{--}6 \text{ bar}$
 $T = 673\text{--}685 \text{ K}$
 $\phi = 0.6$
 $Re_D = 39\text{--}120 \text{ k}$



DME jet flames

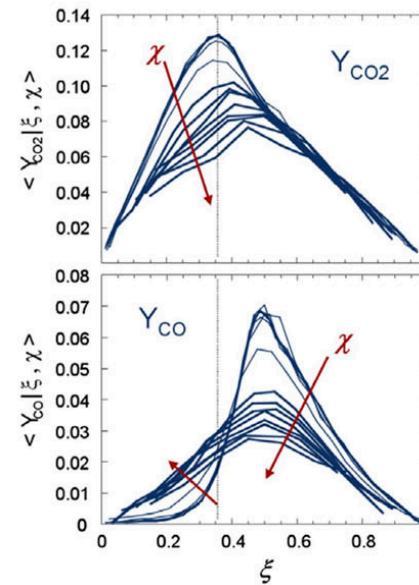
Dual resolution spectrometer to cover the range of shifts



(a)



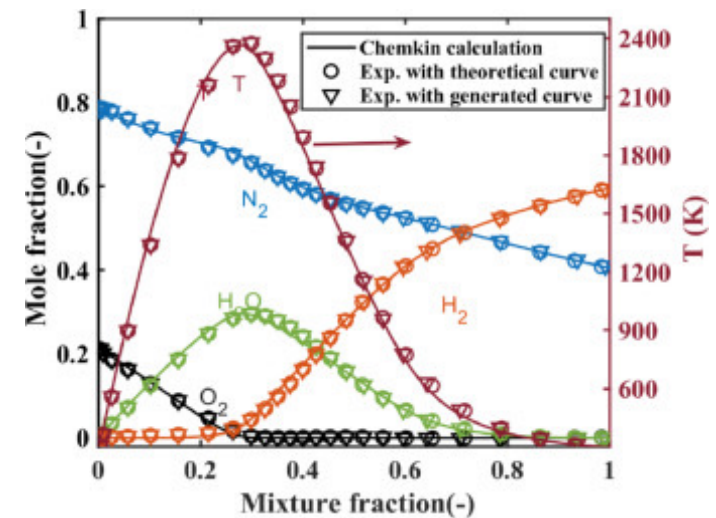
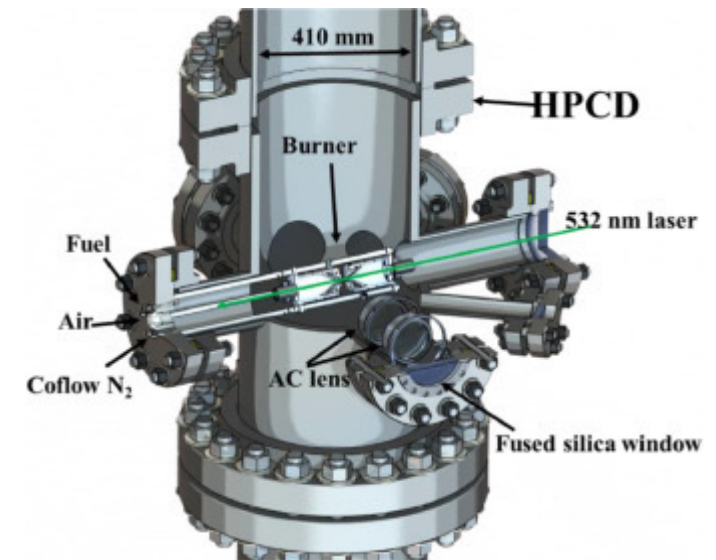
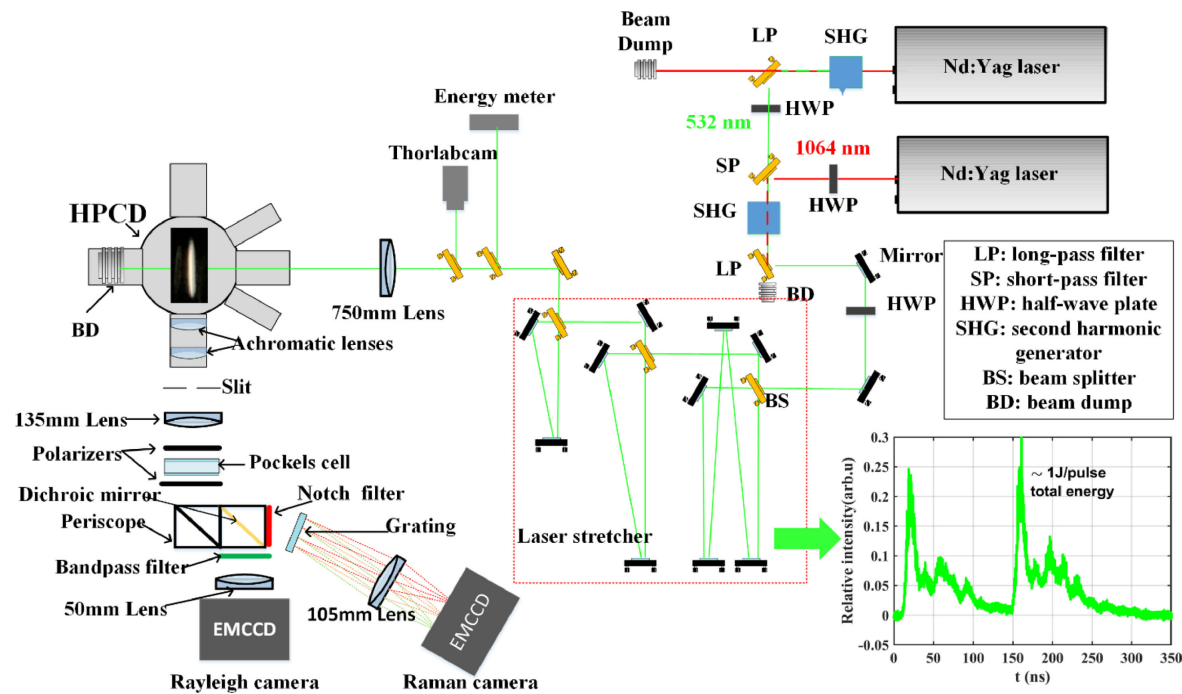
(b)



(c)

[G. Magnotti and R. S. Barlow, Proc. Combust. Inst. **36**, 4477 \(2017\).](#)
[F. Fuest, G. Magnotti, R.S. Barlow, J.A. Sutton, Proc. Comb. Inst., 35 \(2015\)](#)

New fuels investigated with Raman/Rayleigh



[Tang, H., Yang, C., Wang, G., Guiberti, T. F. and Magnotti, G., Comb. Flame 237:111840 \(2022\).](#)

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Summary: Scattering techniques

- Rayleigh/Mie scattering:
 - Simplest technique
 - Dominated by potential particle contamination in the flow
 - Useful for determining temperatures if composition is known
 - Low cross section (signal)
- Raman scattering:
 - Capable of detecting several specific species in one signal
 - Low cross section: requires high laser energies
 - Sensitive to interferences from e.g. chemiluminescence, particles, reflections
 - Quantitative measurements can be challenging (calibration with temperature, multiple line interference)

Questions

- Can I measure temperatures inside an engine/gas turbine using Rayleigh scattering?
- Raman seems like a powerful technique. Why is not used more often?
- Can I do fast Raman?
- Can I use Raman to measure OH?
- Can we we use Raman/Rayleigh with sprays?

Questions



Can I measure temperatures inside an engine/gas turbine using Rayleigh scattering?



How do I find out what the characteristic frequencies are?



Raman seems like a powerful technique. Why is not used everywhere?



Can I do fast Raman?



Can I use Raman to measure OH?



Can I use Raman/Rayleigh with sprays?

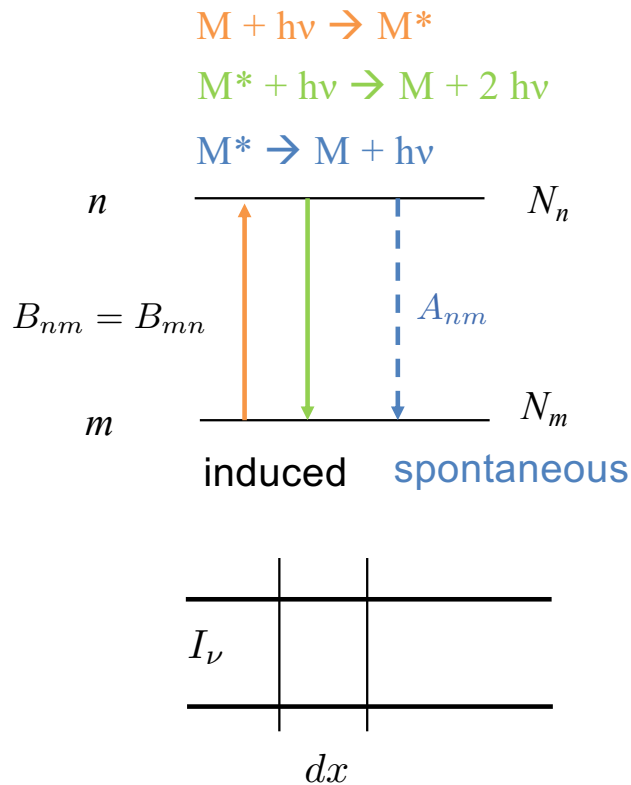


Other questions?

Outline

- Why and how we measure
- Fundamentals of optical diagnostics
 - Light sources and signal connection
 - Detection methods
 - Light-molecule interactions
- Incoherent
 - Scattering and particle velocimetry
 - Rayleigh scattering
 - Raman scattering
 - Absorption
 - Laser induced fluorescence
 - Laser-induced incandescence
- Coherent techniques
 - CARS
 - LIGS

Laser absorption radiation balance



Fraction of laser line overlap

$$\frac{d\rho_\nu}{dx} = N_n A_{nm} f_\nu - (N_m - N_n) \rho_\nu f_\nu B_{nm} \quad \text{change in photons/volume}$$

$$\frac{d\rho_\nu}{dx} = h\nu N_n A_{nm} - h\nu (N_m - N_n) \rho_\nu f_\nu B_{nm}$$

$$\frac{dI_\nu}{dx} = h\nu c \frac{d\rho_\nu}{dx} = N_n A_{nm} f_\nu - h\nu c f_\nu B_{nm} (N_m - N_n) h\nu c \rho_\nu$$

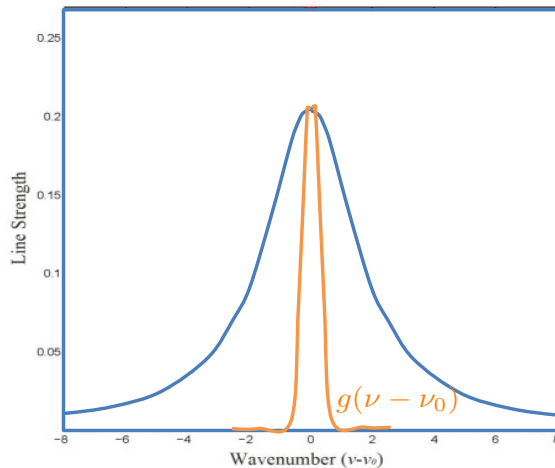
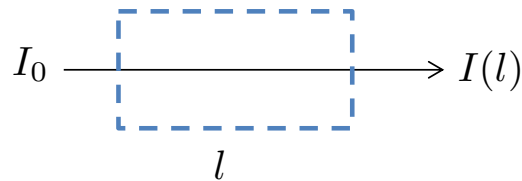
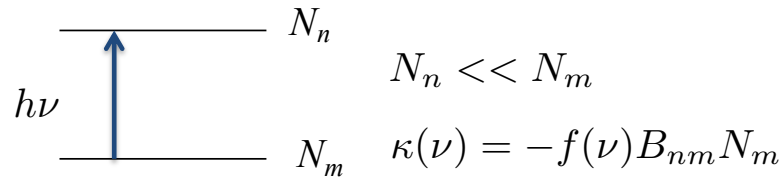
$$\frac{dI_\nu}{dx} = \epsilon(x) - \kappa(x) I_\nu$$

If negligible emission:

$$\frac{d \ln I_\nu}{dx} = -\kappa(x) \quad \text{Beer-Lambert law}$$

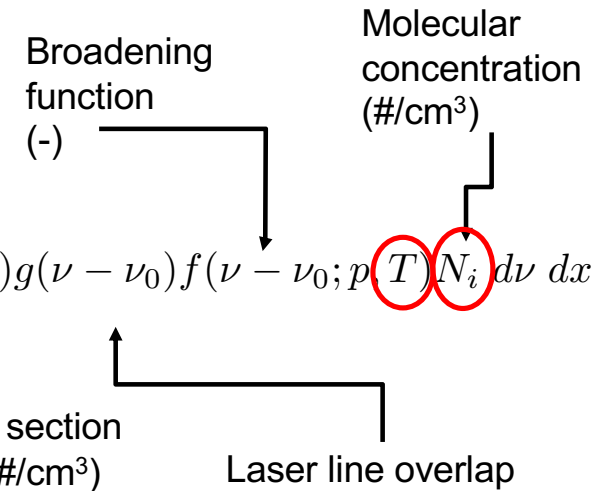
Extinction coefficient

Absorption



Lambert-Beer Law

$$\tau = -\ln \frac{I}{I_0} = \int_0^L \int_0^{\Delta\nu} \kappa(\nu) g(\nu - \nu_0) f(\nu - \nu_0; p(T) N_i) d\nu dx$$



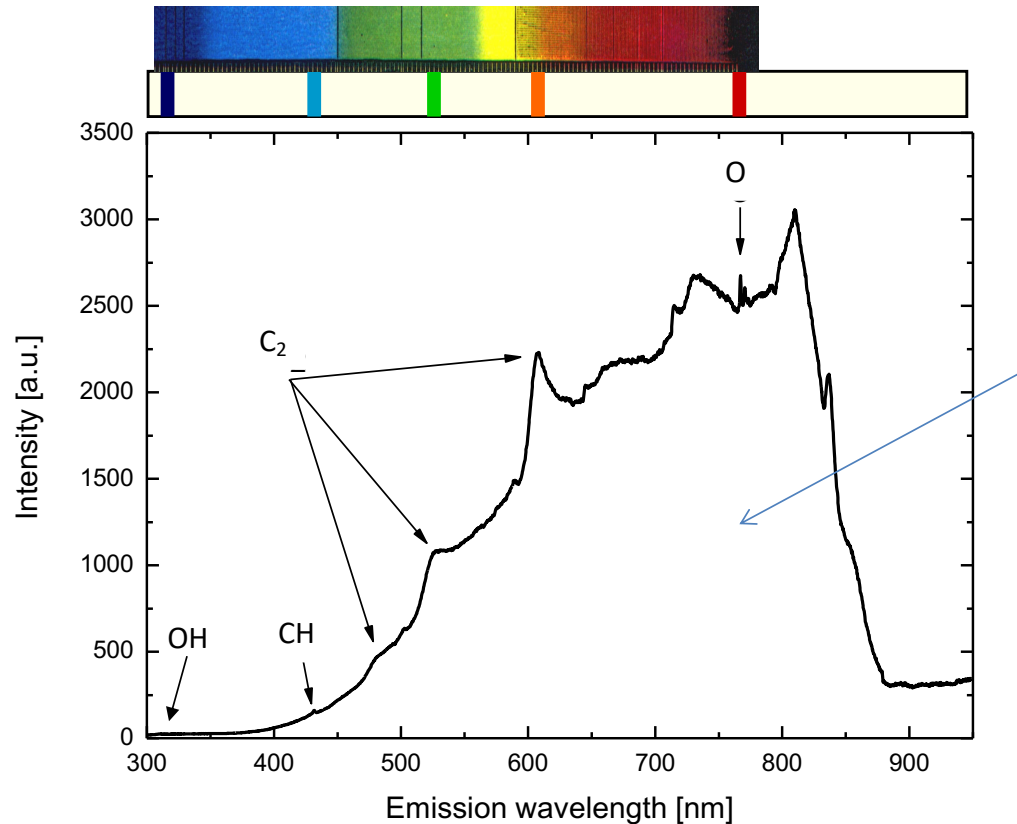
- Inherently line of sight (tomography possible but complex): not spatially resolved
- Lines often weak (need improvement in noise rejection)
- Can work very fast (sub- μs)

Tunable diode lasers:

- Offer very narrow lines over a wide range of useful wavelengths at useful powers
- Can scan a broadened line fast
- Line shape contains temperature information
- Multiple species accessible

Emission and absorption

Flame emission from a candle

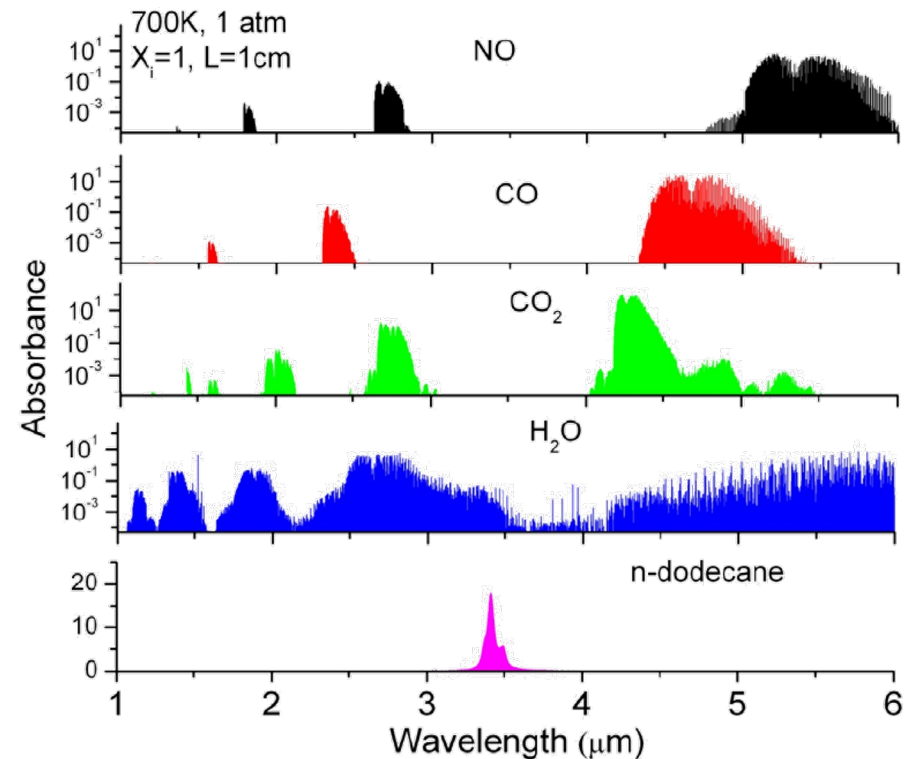


High energies in the visible range: do not overlap with molecular vibration/rotation

Brazil Winter School 2025

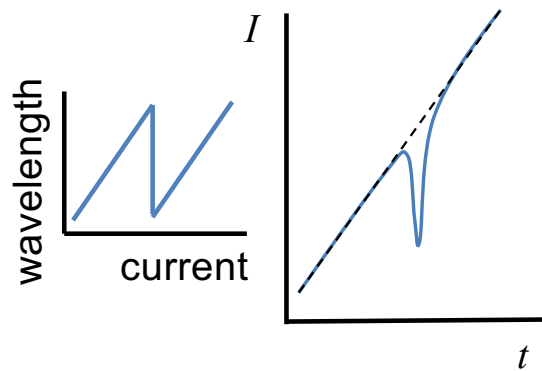
Courtesy Andreas Bültner and
Andreas Brockhinke, Bielefeld

IR absorption spectra from HITRAN



Hanson CISS 2018 Lecture 2

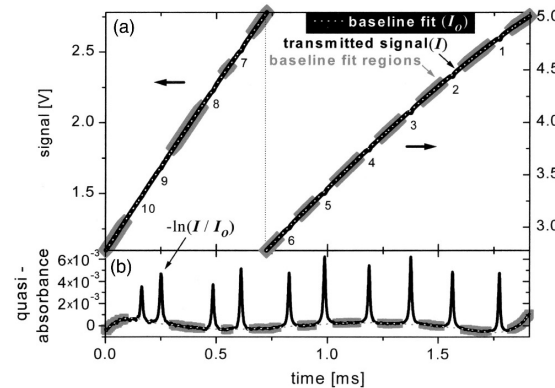
Strategies for removing background



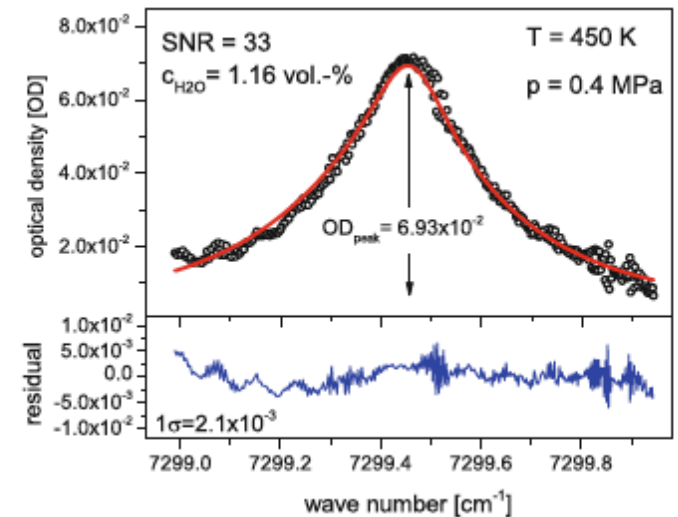
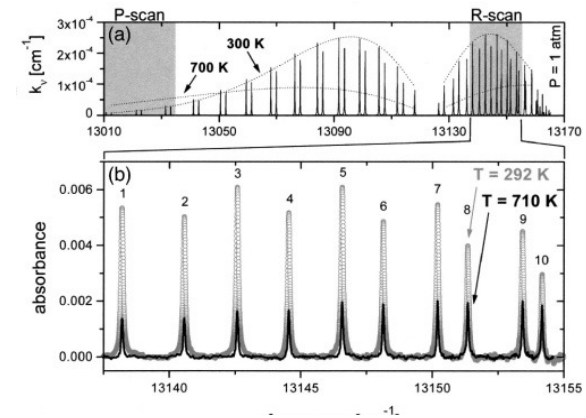
Direct baseline subtraction

Baseline can be noisy
Signal-to-noise ratio can be poor
Complex fitting algorithm

Simple and selective
Simultaneous concentration and line-of-sight temperature

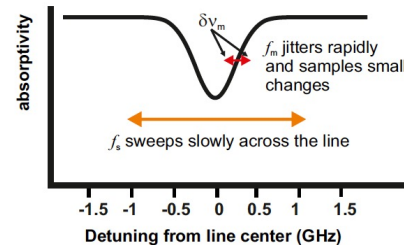
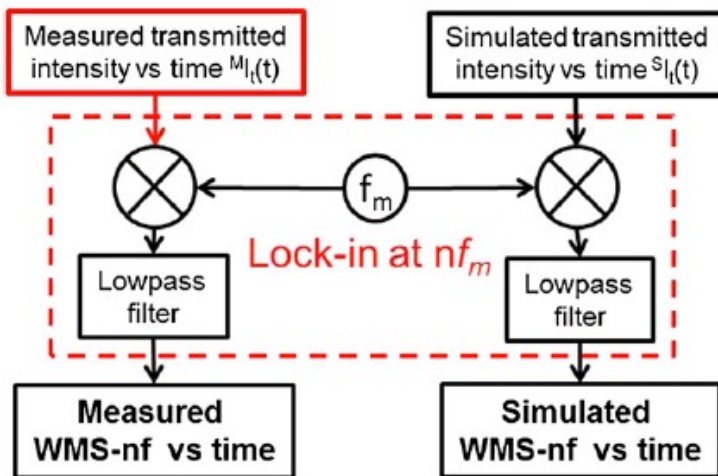
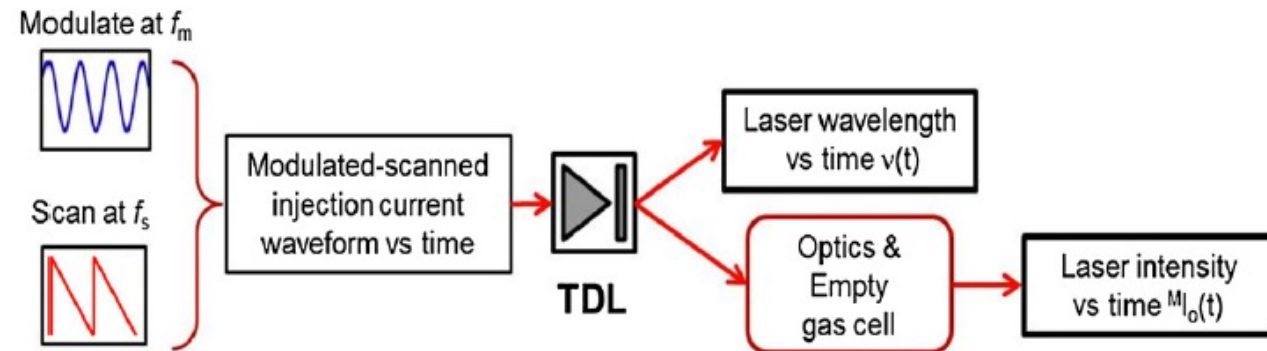


[Sanders et al., Applied Optics, 40,4404-4415 \(2001\)](#)

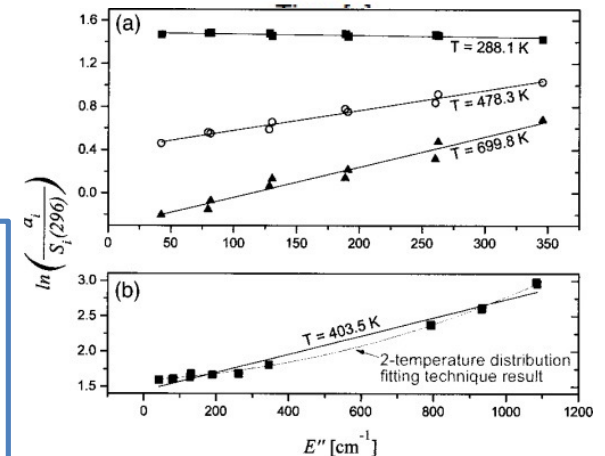
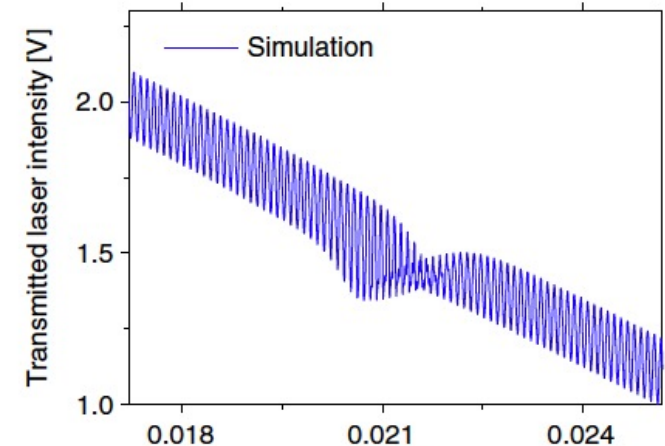


[Witzel et al., Appl. Phys. B \(2012\)](#)

Modulated frequency (and intensity) absorption measurements

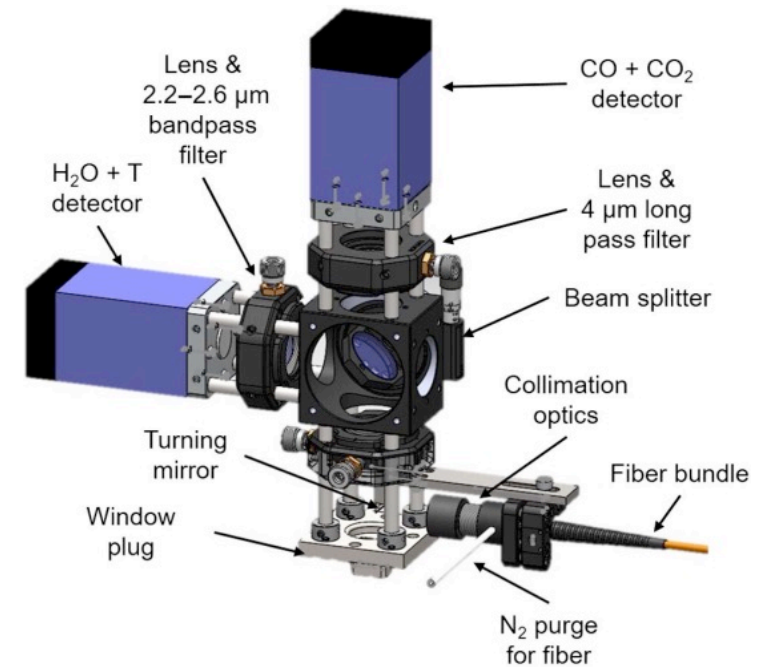
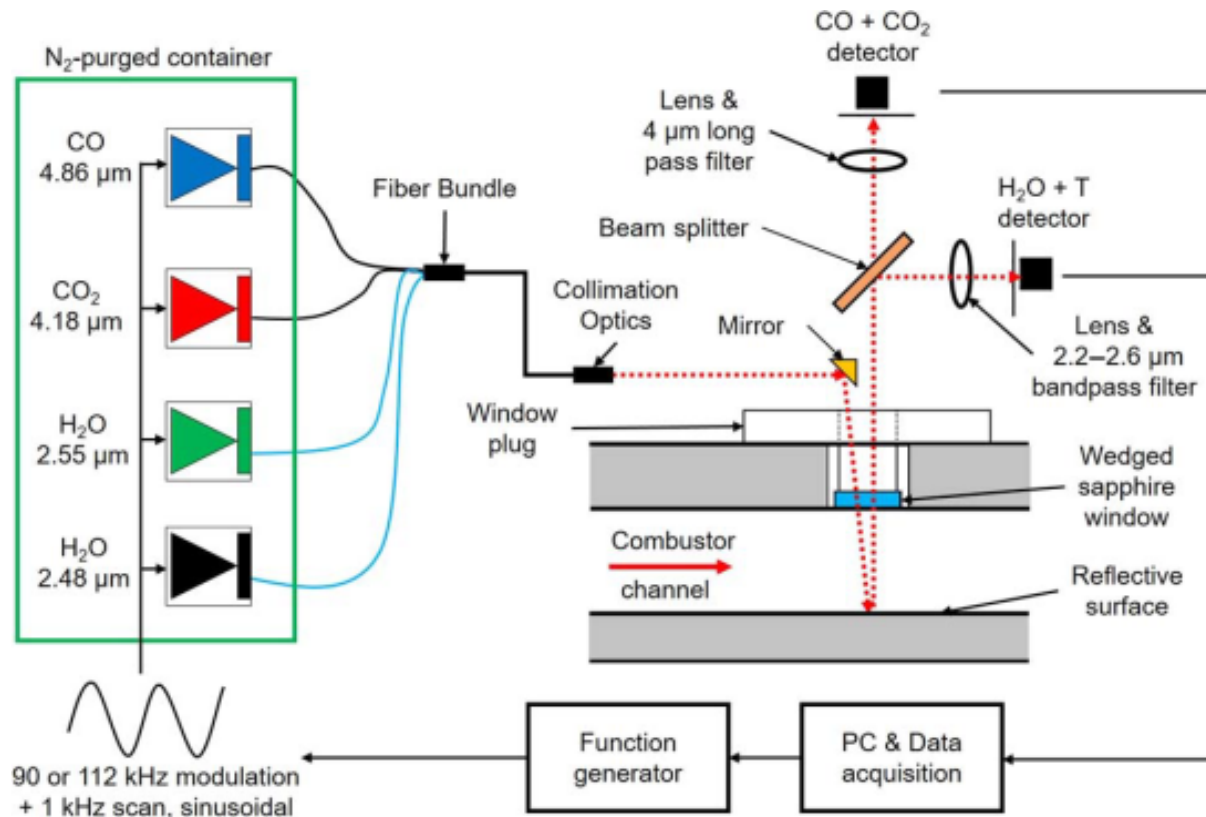


- Lock-in detection at a multiple n of the modulation frequency:
- No need for finding baselines.
- Multiple harmonics: allows canceling of zero-frequency drift



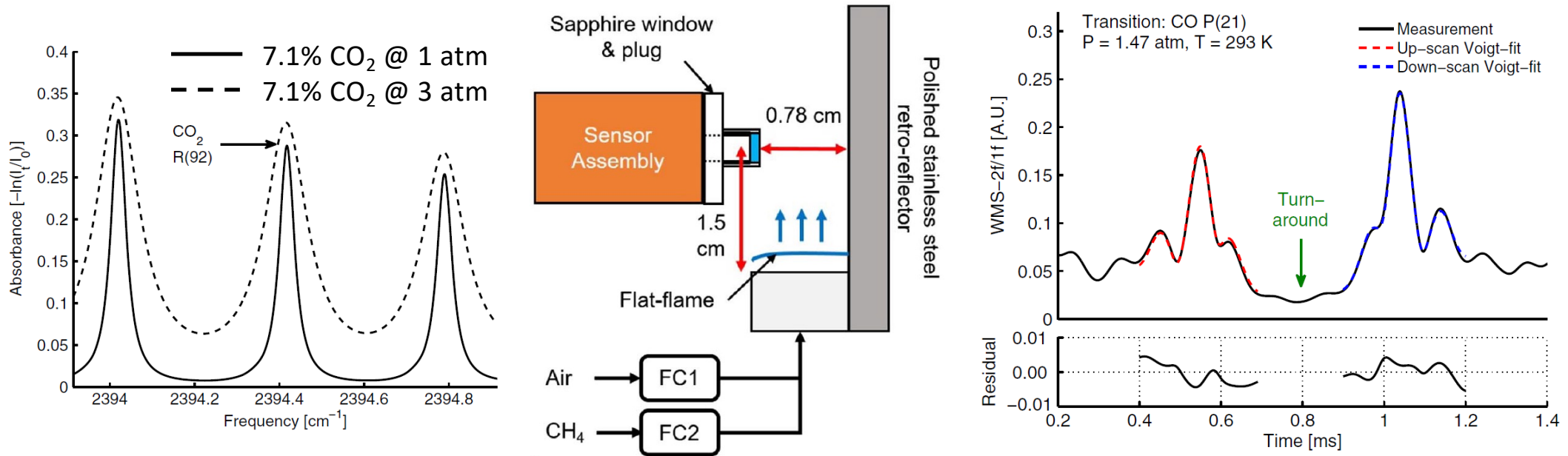
Multi-species: TDLAS of H₂O, CO₂, CO, and temperature

Peng et al., Applied Optics 55 (2014)



Species	ν_0 (cm ⁻¹)	E'' (cm ⁻¹)	S (296 K) (cm ⁻² /atm)
H ₂ O	3920.08	704.2	6.35×10^{-1}
H ₂ O	4029.52	2660.95	1.10×10^{-4}
CO ₂	2394.41	3329.0	7.39×10^{-5}
CO	2055.40	886.9	6.20×10^{-1}

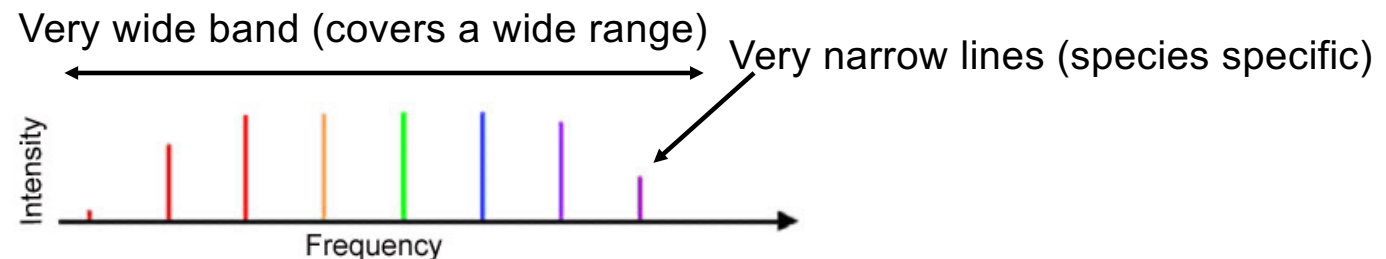
Application example: TDLAS H₂O, CO₂, CO, and temperature



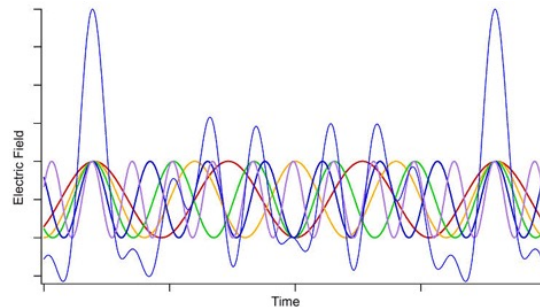
- Simulated absorption spectra of CO₂ in products of $\phi=1.2$ CH₄/air flame (T=1500K, L=15 mm)
- 0.78 cm path length across flat flame products
- Scanned-wavelength-modulation spectroscopy with 2nd harmonic detection and 1st harmonic normalization ([scanned-WMS-2f/1f](#)) suppresses several noise sources
- WMS spectral fitting routine

Frequency combs

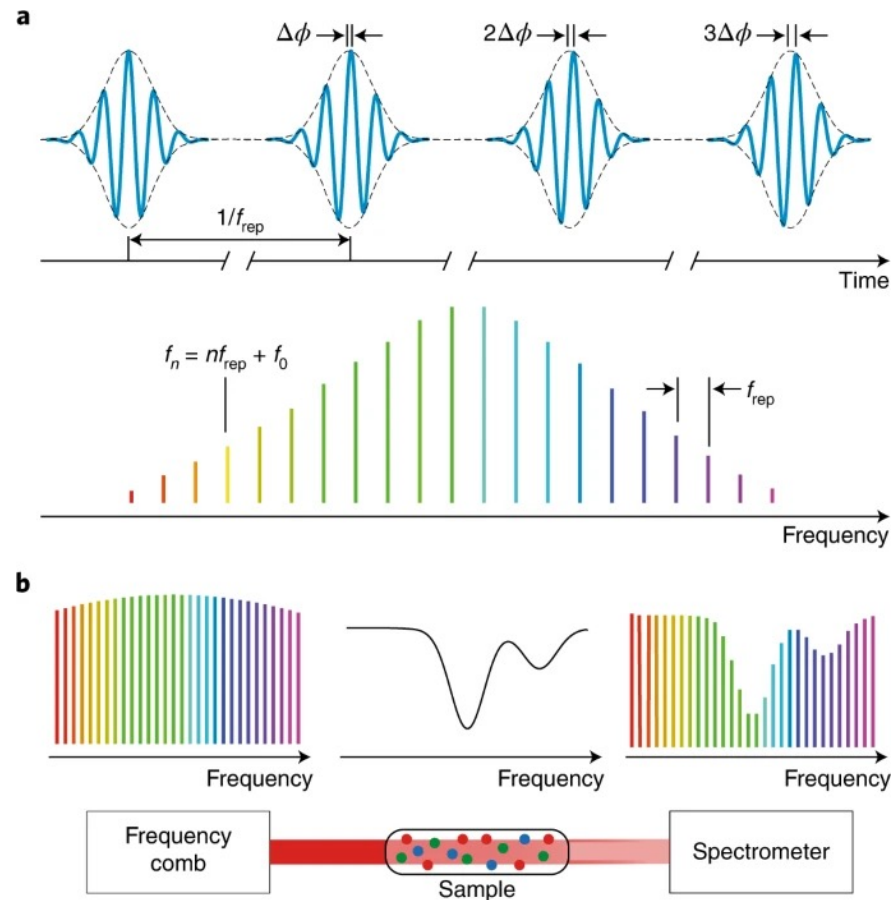
- Tunable lasers have only narrow ranges of wavelengths
- How can this range be improved?



Frequencies generated with known spacing by mode (phase) locking two lasers at precise intervals

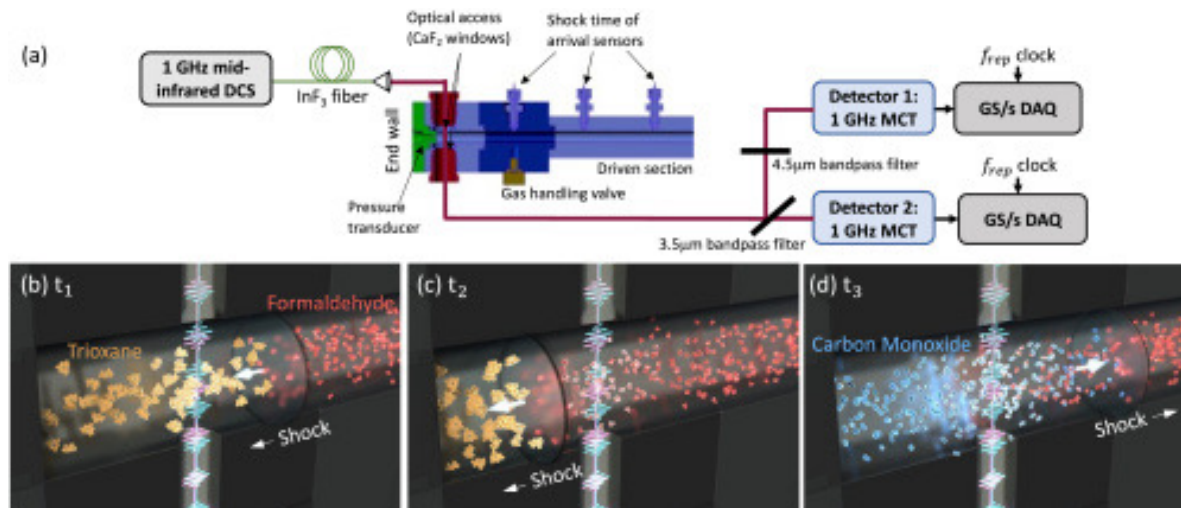


Principle of operation

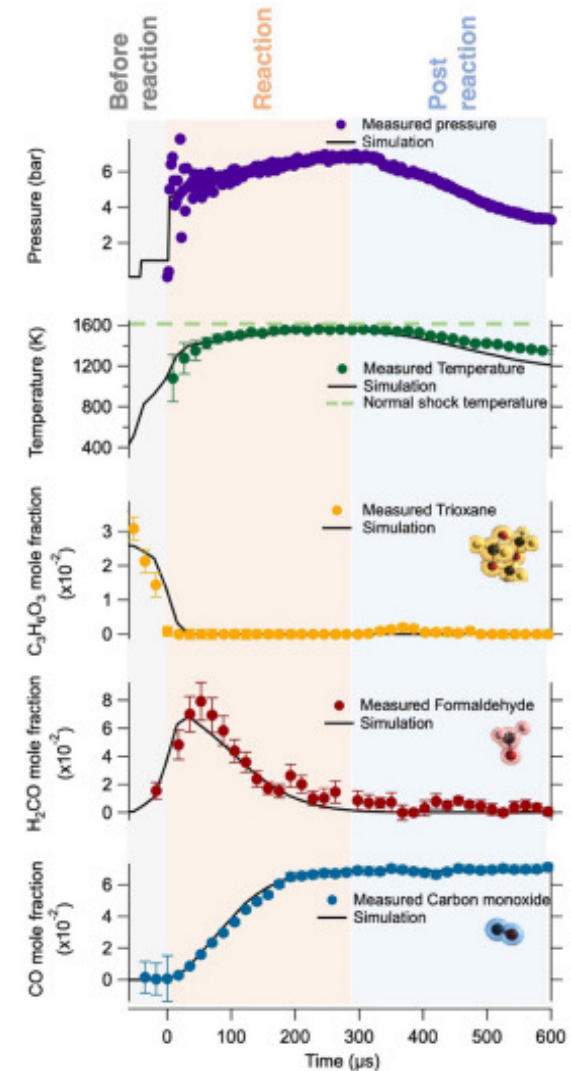


Piquet & Hänsch, Nature, 2019

Example frequency comb application



Hoghooghi et al. GHz repetition rate mid-infrared frequency comb spectroscopy of fast chemical reactions, Optica (2024)



Databases

Original, created for
atmospheric/astronomy studies

<https://hitran.org>

HITRANonline [Login](#) [Register](#)

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The HITRAN Database

HITRAN is an acronym for *high-resolution transmission* molecular absorption database. HITRAN is a compilation of spectroscopic parameters that a variety of computer codes use to predict and simulate the transmission and emission of light in the atmosphere. HITRAN data is predominantly used in atmospheric science and astrophysics, but it has many applications throughout science, industry, and technology.

The HITRAN project is directed from the [Center for Astrophysics | Harvard & Smithsonian](#).

News

- Jul 2025 | [HITRAN2024 edition will be released in the Fall of 2025](#)
- Jun 2025 | [37000 users milestone](#)
- Aug 2024 | [Call for papers for the HITRAN special issue in JQSRT \(closing May 1, 2025\). ★](#)
- Dec 2023 | [HITRAN now includes water](#)

Incorporates hitran, and extends
it to high temperatures

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What's New at SpectralCalc!

- [HITRAN 2020](#) — All HITRAN gases (1-55) now available.
- [HITRAN 2020](#) — The 2020 HITRAN line data is now available.
- [HITRAN 2016](#) — The 2016 HITRAN line data is now available.
- [Water Continuum](#) — Model updated to use coefficients derived from 2.5.2

Gas-Cell Simulator
Atmospheric Paths

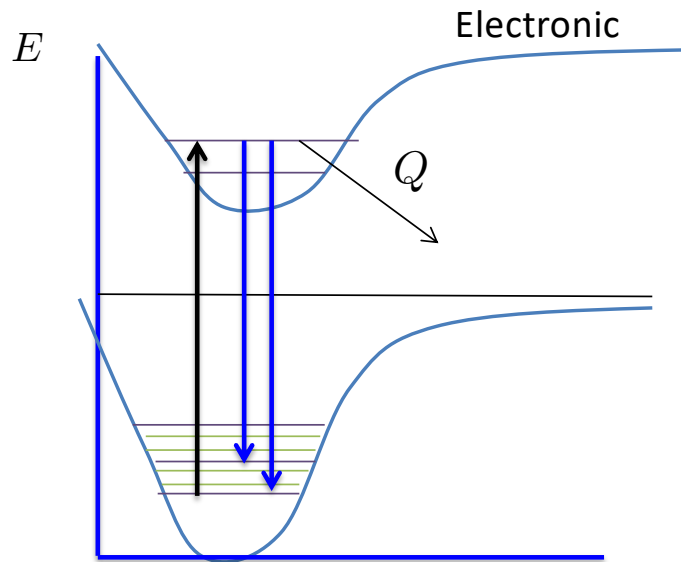
Summary: Absorption techniques

- Can be very selective sensitive (multi-pass cell, modulation spectroscopy)
- Can get mean temperature/pressure from multiple line shapes
- Systems can be very compact; fiber optics
- Line of sight technique; temperature and composition ideally uniform or symmetric along the path length
- Sensitive to moisture, temperature
- Special windows (KBr) required
- Emerging technique: frequency combs opening up new ranges of wavelengths for simultaneous detection of multiple species
- Applications: fast processes where spatial resolution is not important or symmetry present

Outline

- Why and how we measure
- Fundamentals of optical diagnostics
 - Light sources and signal connection
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 - Light-molecule interactions
- Incoherent
 - Scattering and particle velocimetry
 - Rayleigh scattering
 - Raman scattering
 - Absorption
 - Laser induced fluorescence
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 - CARS
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Laser Induced Fluorescence



$$I_{LIF} = KI_0 N(p, T) f_{\nu, i}(T) B_{ik} \Gamma(p, T) \phi$$

Boltzmann
fraction

Line overlap

Quantum
yield

$$\phi = \frac{A_{ki}}{\sum_j A_{kj} + Q_k + P_k}$$

Two (or multi)-line LIF

$$\frac{I_{LIF, \nu_1}}{I_{LIF, \nu_2}} \propto \exp\left[-\frac{\Delta\epsilon_{12}}{k_B T}\right]$$

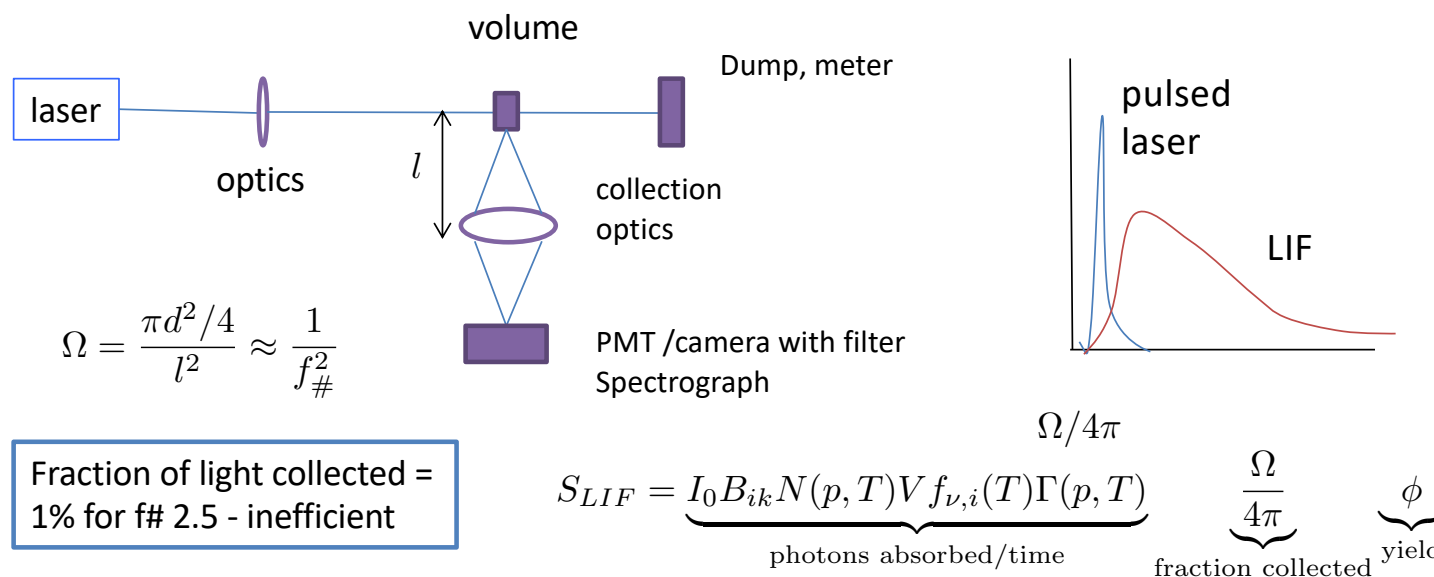
- **High energy gap**, so wavelengths required are in the UV, but **strong signal** sometimes
- Signal returns at a different wavelength than pump: can **separate spectrally**
- Only works for **molecules with accessible levels** in the visible and near UV (OH, CO, NO, CH₂O accessible convenient, HCO, CH, H very low signal)
- Signal strongly affected by p, T and **quenching**: collisional corrections required
- Imaging depends on **good knowledge of specific line behaviour**
- Temperature dependence: used to **obtain T** for known lines

A few important combustion species detected by LIF

Molecule	Excitation	Detection
OH	~283 nm, ~314 nm	306-320 nm
NO	~226 nm	236-280 nm
CH	~314, ~387, ~431 nm	various 314-460 nm
CO	~230 nm (2 photon)	~484 nm
CH ₂ O	~355	360-550 nm
HCO	~258 nm	276-284 nm
NO	226 nm	250 nm
Acetone, Propanone	225-320 nm	Broadband, ~350 nm
PAH	<300	broadband
Toluene	240-270	300
2-pentanone, Biacetyl	250-320	250-320

- References:
 - Applied Combustion Diagnostics, Kohse-Höinghaus and Jeffries, Eds., Chapter 2, 2002
 - <https://lifsim.com>: Modular laser induced fluorescence code for common combustion species.

Typical LIF experimental setup

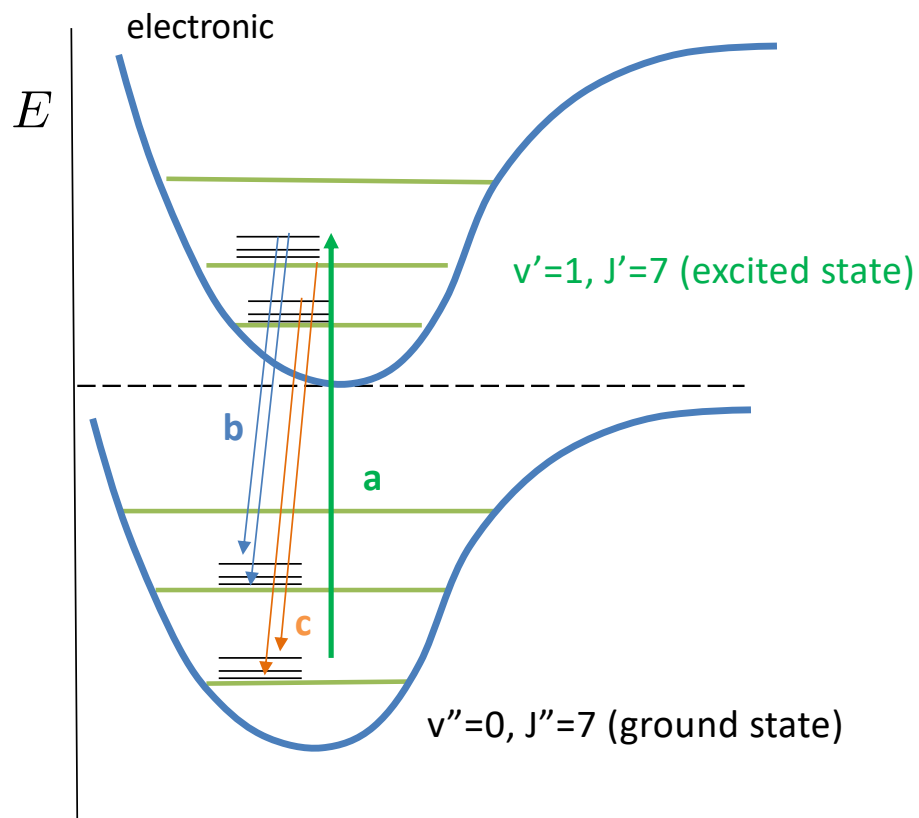


$$\phi = \frac{A_{ki}}{\sum_j A_{kj} + Q_k + P_k}$$

Usually a small number at normal pressures

Powers required of the order of kW: ok for pulsed lasers: 10 mJ/10 ns = 10^5 - 10^6 W
 Saturation of upper electronic level – loss of linearity with power: need to know dependence
 For some molecules, ppm values achievable.
 Quantitative LIF an order of magnitude more challenging than qualitative.

Electronic transitions: OH laser induced fluorescence (LIF)



Adapted from Linne CISS 2016

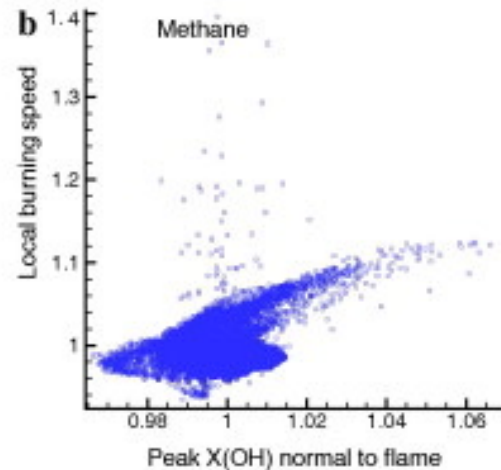
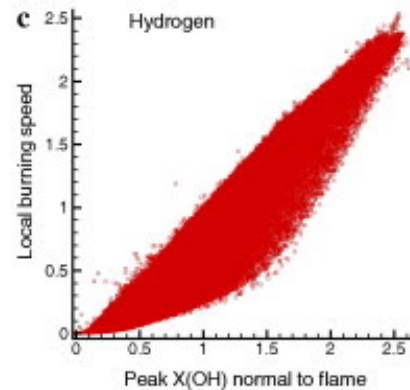
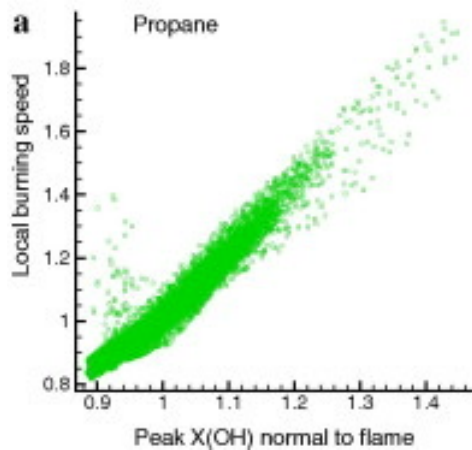
- Typical strategy for OH LIF or PLIF imaging
- **Excitation** (molecule absorbs, **a**, usually single line)

$$Q_1(7) \ A^2\Sigma^+ \leftarrow X^2\Pi(1,0) \quad (283.2 \text{ nm})$$
- Most molecules relax via **quenching collisions** (process **b**)
- **Detection** (molecule emits after rotational and vibrational energy transfer; many different transitions, process **c**)

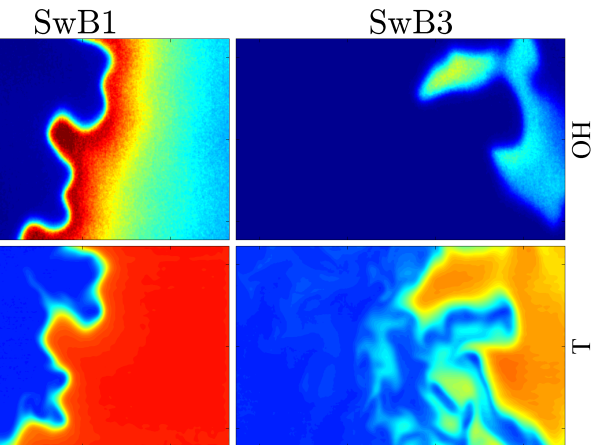
$$A^2\Sigma^+ \rightarrow X^2\Pi(1,1),(0,0) \quad (305\text{-}320 \text{ nm})$$

- To make **quantitative** measurements, we need to know (among other things):
 - population of the ground state ($v''=0, J''=7$)
 - efficiency of excitation and fluorescence
 - collisional quenching rate

OH LIF: frequent marker of location of flame



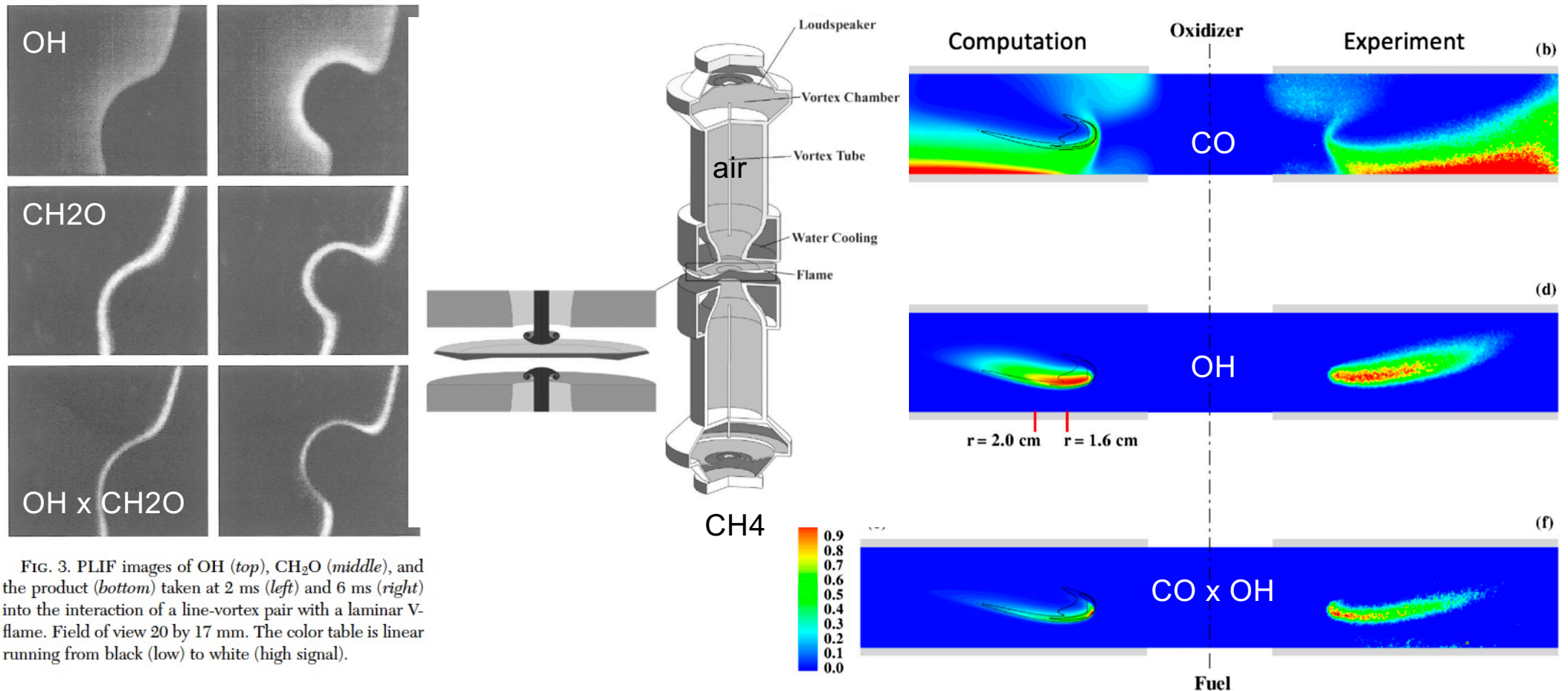
[Bell, J. B. et al., PCI, 31:1 \(2007\)](#)



[Kamal et al., Proc. Comb. Inst. 36 \(2017\) 1957–1965](#)

- Coincides with reaction zone within $\sim 50 \mu\text{m}$
- Correlates with reaction rate for some reactant mixtures

Reaction rate imaging: $\text{CH}_2\text{O} + \text{OH}$, and $\text{CO} + \text{OH}$



Paul, P. H. and Najm, H. N. PCI (1998) 27:43-50

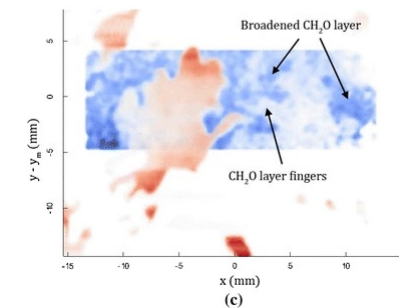
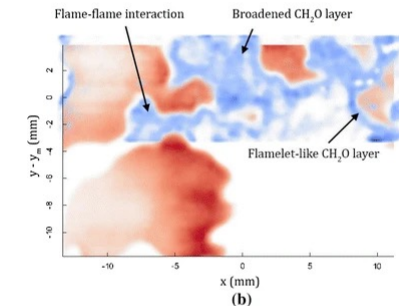
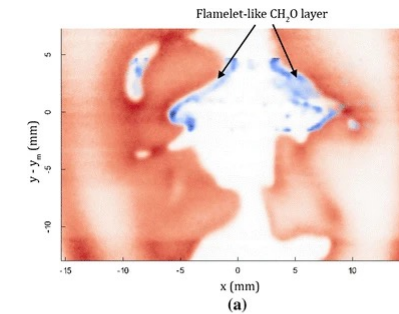
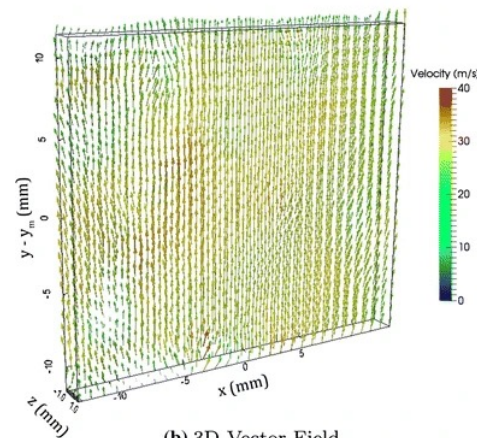
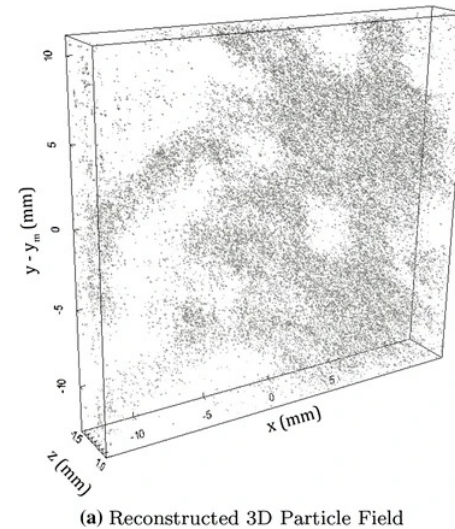
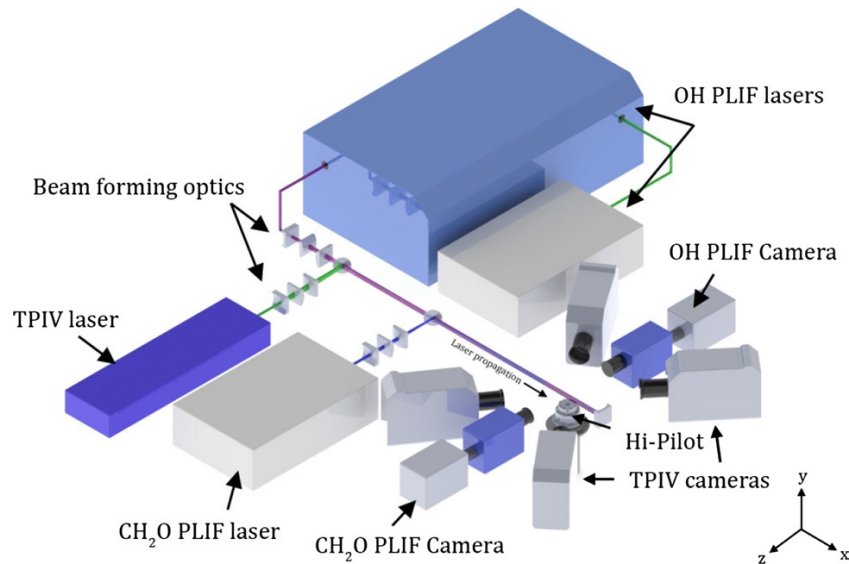
Brazil Winter School 2025

Amantini *et al.*, Combust. Flame 147, 133 (2006)



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Simultaneous OH and CH₂O LIF (non-quantitative)

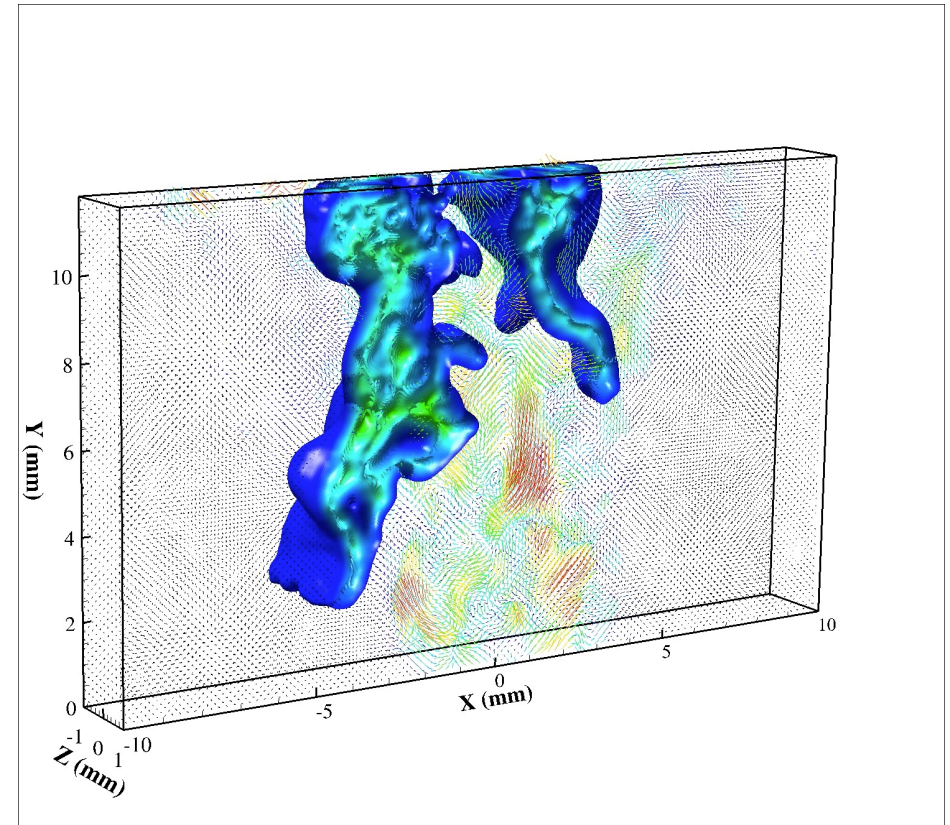
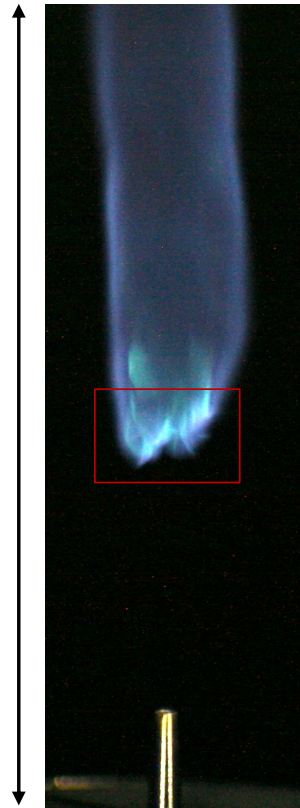


Osborne, J.R., Ramji, S.A., Carter, C.D. *et al.* Simultaneous 10 kHz TPIV, OH PLIF, and CH₂O PLIF measurements of turbulent flame structure and dynamics. *Exp Fluids* **57**, 65 (2016).

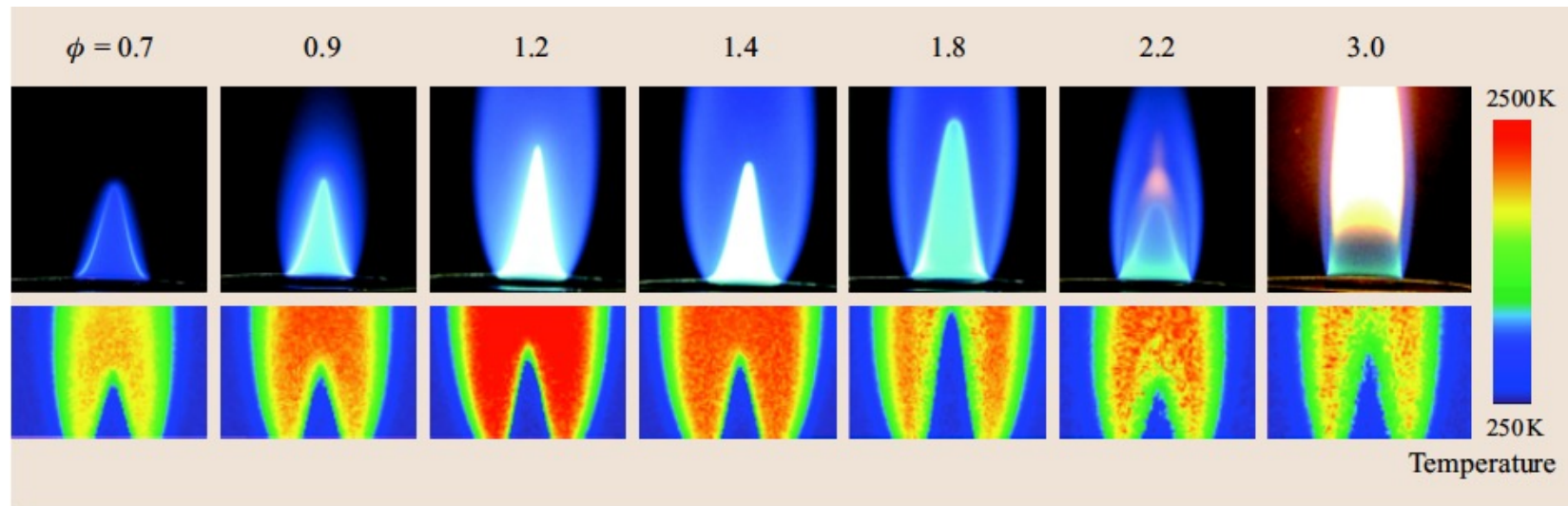
High-Speed 3D OH LIF and Velocity at Base of Lifted Jet Flame

Lifted Dimethyl
Ether (DME)/Air
Jet Flame

102 mm



NO TLIF



W.G. Bessler, C. Schulz: Quantitative multi-line NO LIF
temperature imaging, Appl. Phys. B 78, 519–533 (2004)

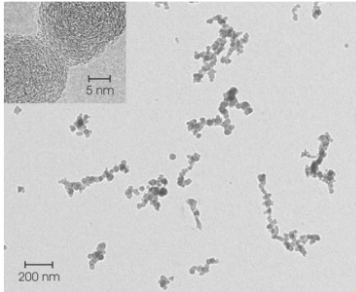
Summary LIF

- Workhorse of combustion: species of interest accessible in the UV: OH, CH, HCO, CH₂O, NO, NH₃, aromatics, ketones
- Cross section much larger than Raman
- Informs flame and combustion structure
- Quantitative measurements one order of magnitude harder than qualitative
- Easy to combine with velocity measurements: great insights into flow-flame interactions
- Limited by laser power to illuminate volumes

Outline

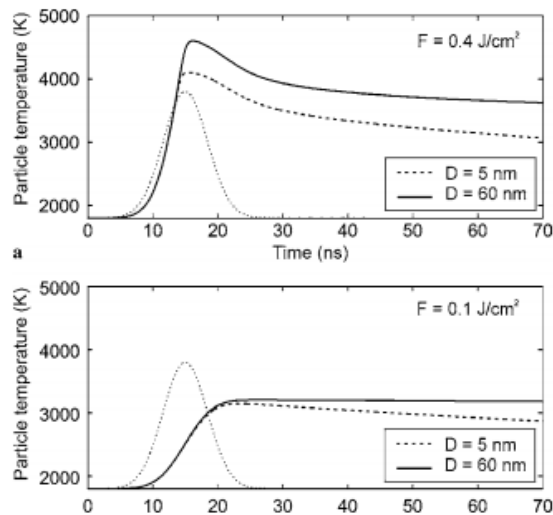
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Laser-Induced Incandescence

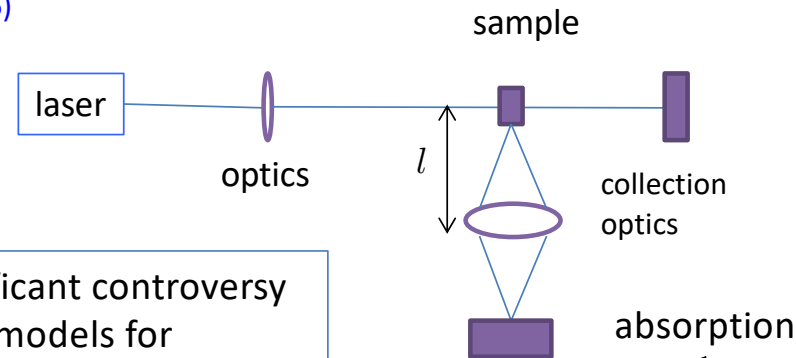


Soot: black body particles
Absorb light in the Rayleigh regime (small particles)
Heat up instantaneously to very high (4000 K) temperatures
Emit incoherent radiation as black bodies

Soot TEM
(Shaddix, 1996)



Bladh et al., Appl. Phys. B, 2008



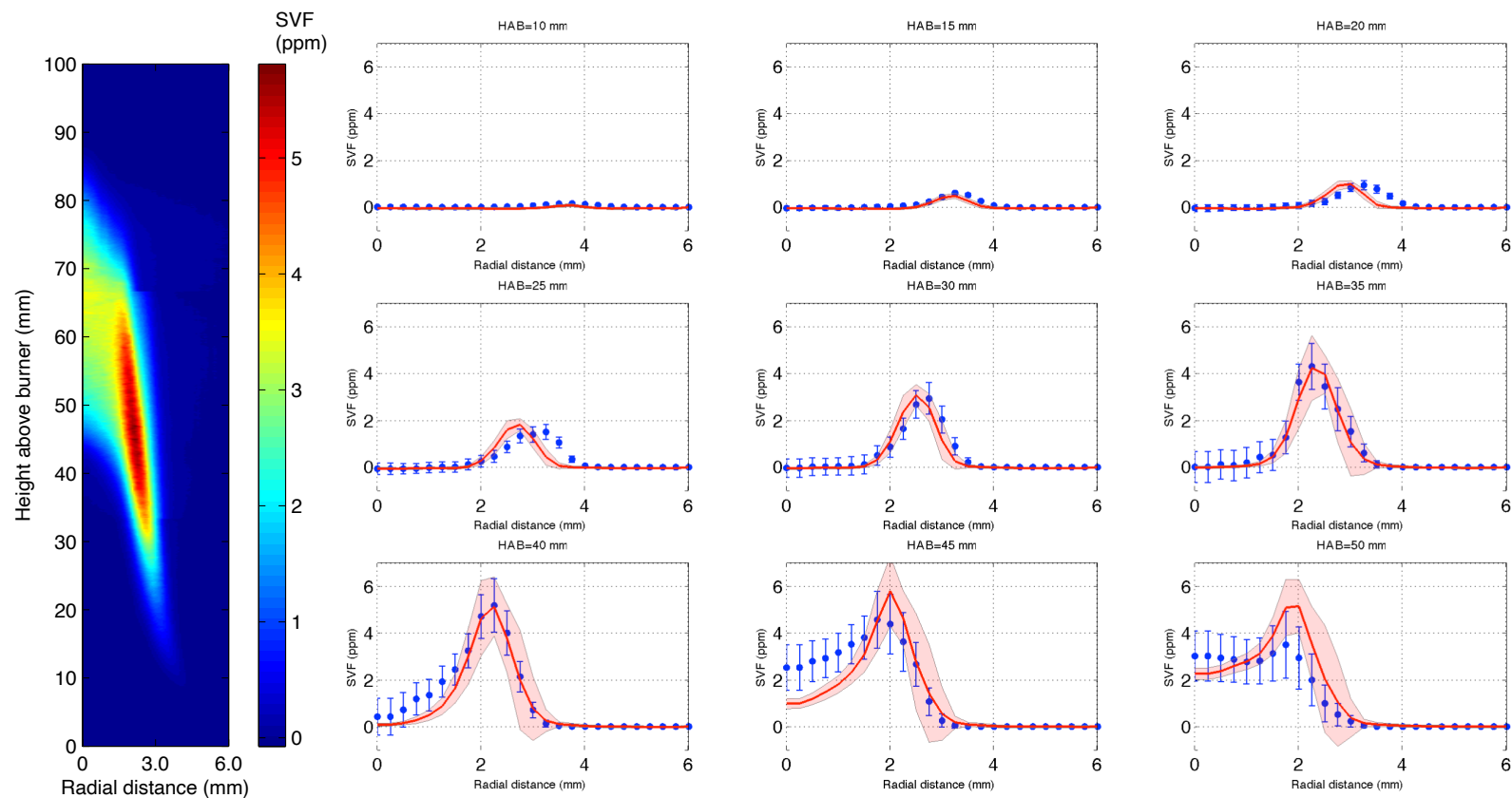
Significant controversy
over models for
cooling!!

$$S = \frac{12\pi c^2 h E(m)}{\lambda^6 [\exp(-\frac{hc}{k\lambda T}) - 1]} \eta(\lambda) f_v$$

Black body

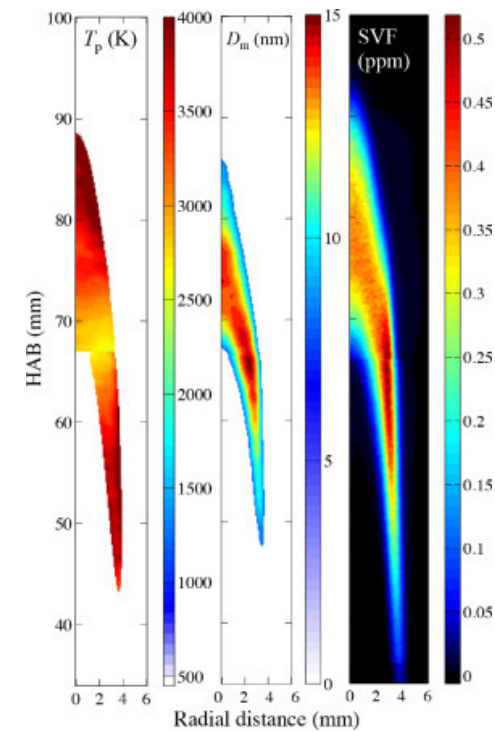
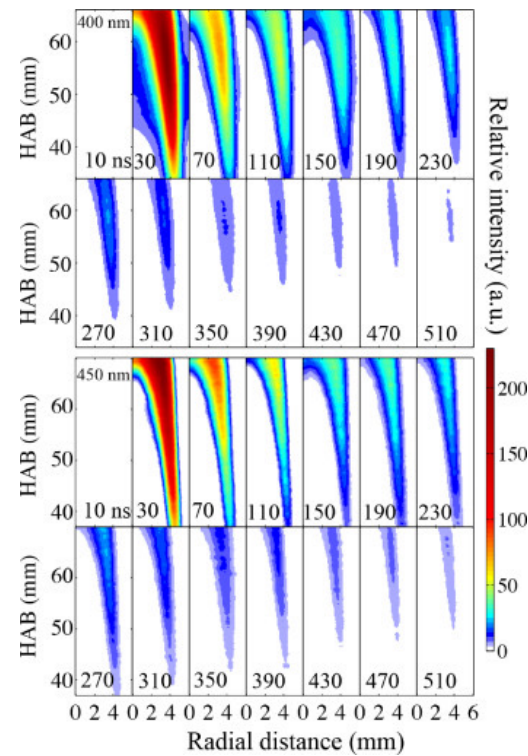
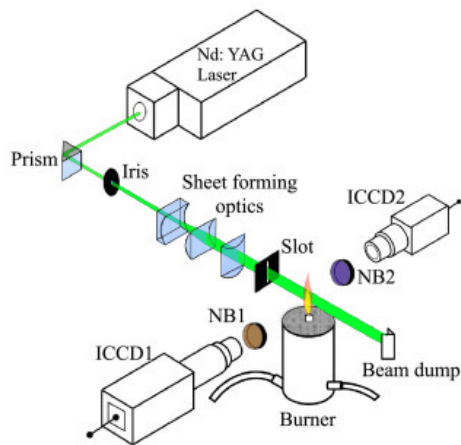
volume fraction

Propane flame soot volume fraction: LII vs extinction



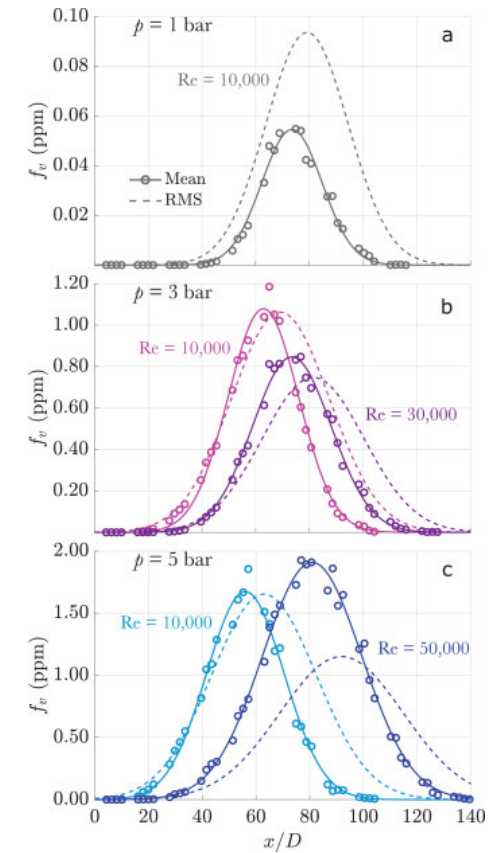
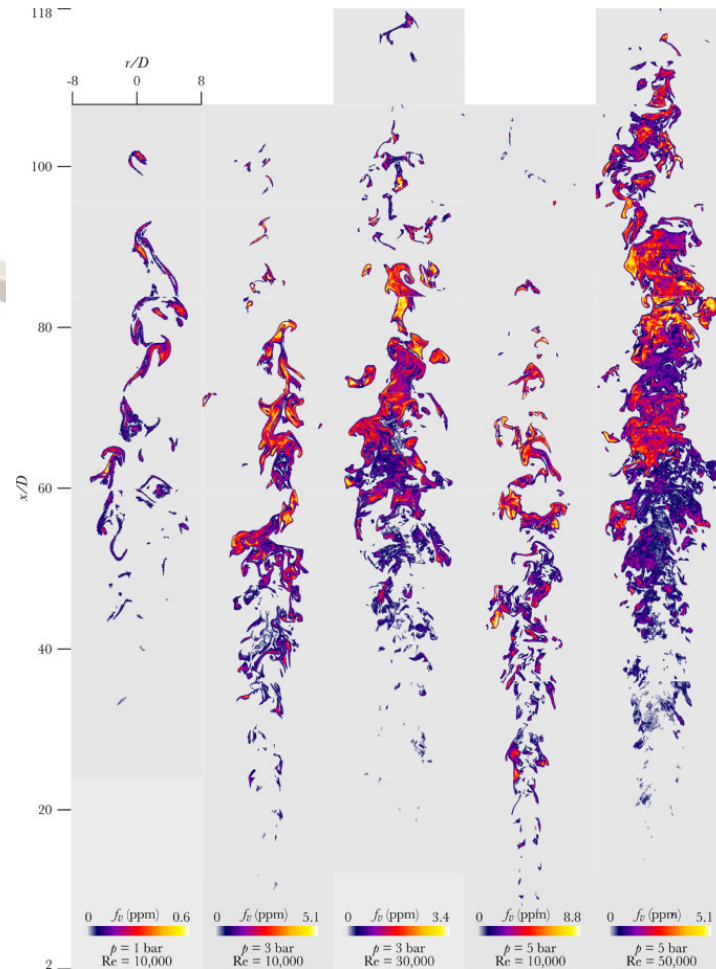
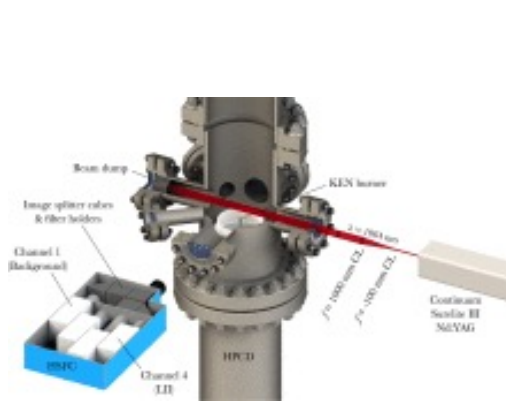
B. Tian, Y. Gao, S. Hochgreb, APB 120, 2015

LII and particle sizing using two color pyrometry



[B. Tian, et al., Proc. Comb. Inst. 39 \(2023\)](#)

Effects of pressure and turbulence on soot formation



Boyette et al., C&F 2021

<https://doi.org/10.1016/j.combustflame.2021.01.013>

Brazil Winter School 2025

Questions



Why is LIF so popular?



What are the problems with making LIF quantitative?



How do I know whether a species will fluoresce?



How do I correct for LIF quenching?



Do all particles incandesce?

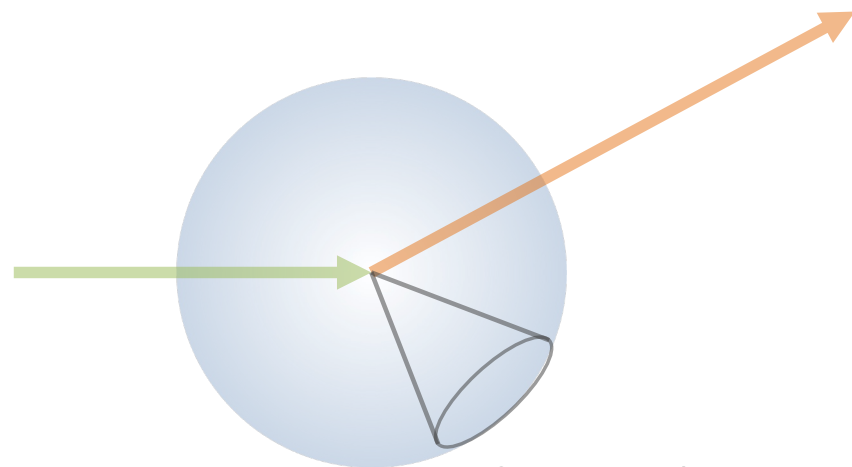


Is LII always proportional to volume fraction?

Outline

- Why and how we measure
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 - LIGS

Coherent (non-linear) processes



Scattering/emission

$$S \propto \frac{K}{r^2}$$

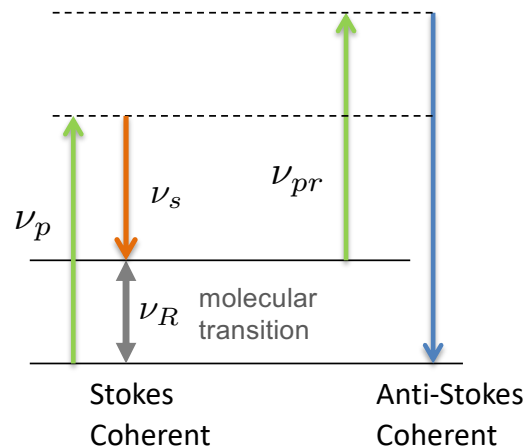
Coherent beam:

- Signal decreases very little with distance
- Can be used to exclude **noise** from collection solid angle by taking the beam over long distances from the sample location

Mission:

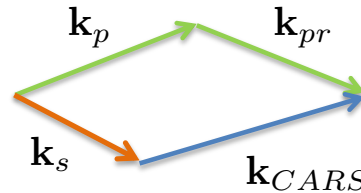
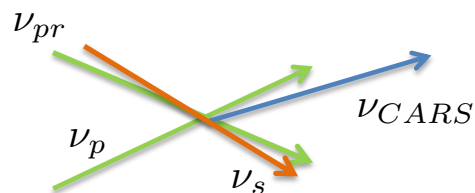
- Generate coherent beam that contains the wavelengths/intensities encoding the quantities of interest

Coherent Anti-Stokes Raman Scattering (CARS)

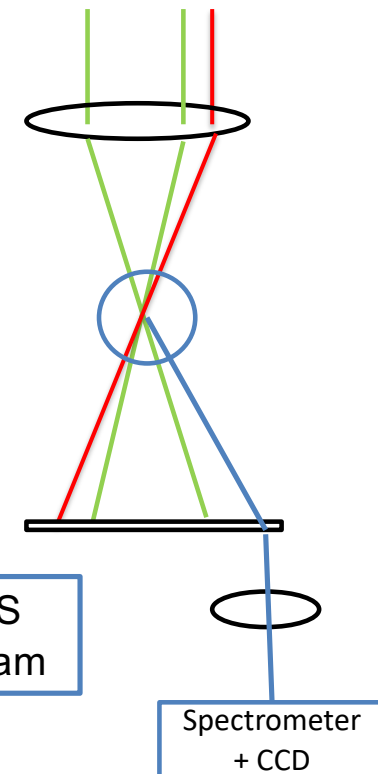


$$\nu_{CARS} = 2\nu_{pr} - \nu_s$$

$$I_{CARS} \propto N^2 I_p I_{pr} I_s$$

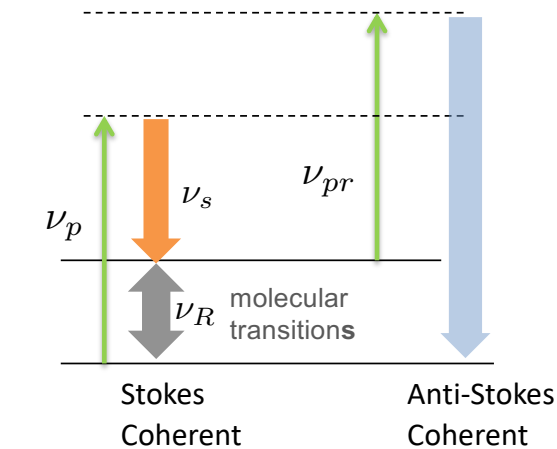


Scan k_s to get CARS
Signal is a laser beam



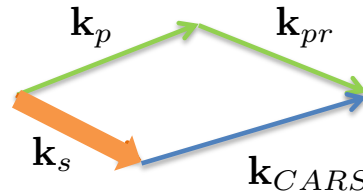
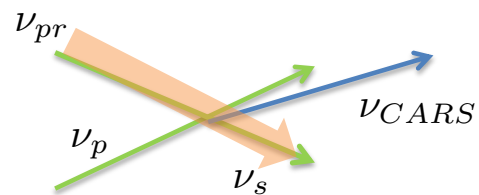
Why use such a complicated method?
There are no IR pulsed lasers to reach molecular transition directly
Method amplifies the weak stokes signal

Broadband Coherent Anti-Stokes Raman Scattering (CARS)

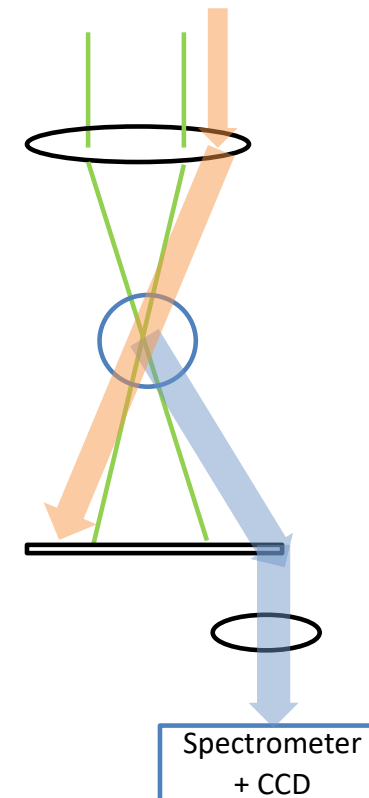


$$\nu_{CARS} = 2\nu_{pr} - \nu_s$$

$$I_{CARS} \propto N^2 I_p I_{pr} I_s$$



$\mathbf{k}_s$ broadband to get CARS broadband
Signal is a laser beam



ns-CARS pros and cons

Spectral selectivity: select the wavelength range that best suits the transition without interference from other species (but limited by availability of lasers).

Laser-like directional signal: coherent light, very high directionality, detection can be done over a small solid angle, detector can be placed far away from the sample.

Good spatial resolution: Beam crossing region has high selectivity (typ. <500 μm)

Species and temperature: like LIF, can be used both for concentration and temperature measurements.

Complex set up: Demands precise (single mode, long coherence length) laser sources (at least two), *phase matching* can be tricky, especially with enclosures. **Expensive.**

Non-resonant background (NRB): Modeling the signal can be onerous, especially because of the non-resonant background. Requires specialist knowledge.

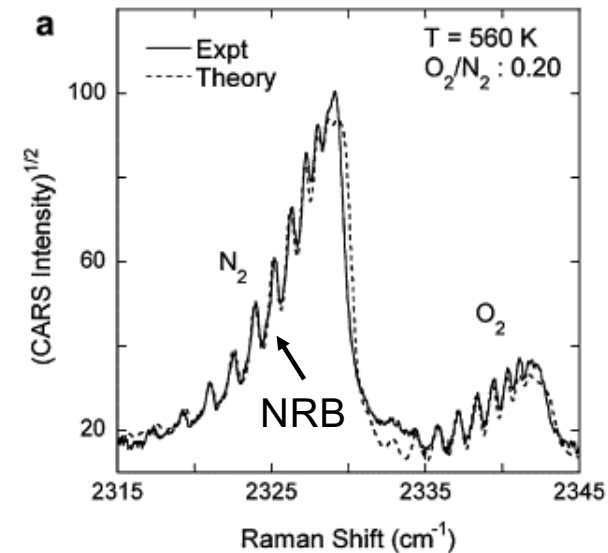
Recent developments:

rotational CARS

broadband CARS

femtosecond/picosecond CARS

2D CARS



[Roy, S., J.R. Gord, A.K. Patnaik, Prog. Energy Combust. Sci. 36 \(2010\) 280–306 doi:10.1016/j.pecs.2009.11.001.](#)

Dual pump fs/ps-CARS

Very short pulses: very high peak power

Femtosecond (fs): 10^{-15} s

Picosecond (ps): 10^{-12} s

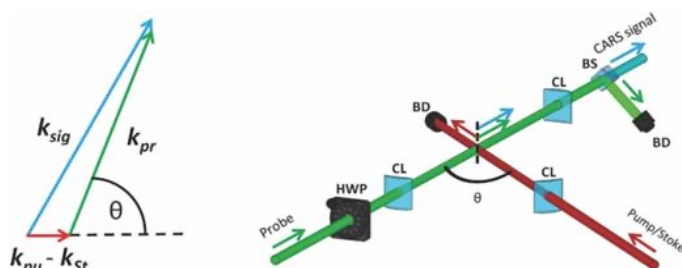
(nanojoules/pulse) = 10^{-9} J / 10^{-12} s = kW, 10^{-9} J / 10^{-15} s = MW

Very short pulses and uncertainty principle: fs pump is broadband

$$h\Delta\nu \cdot \Delta t > \frac{h}{4\pi} \quad \Delta\nu \cdot \Delta t > 0.1$$

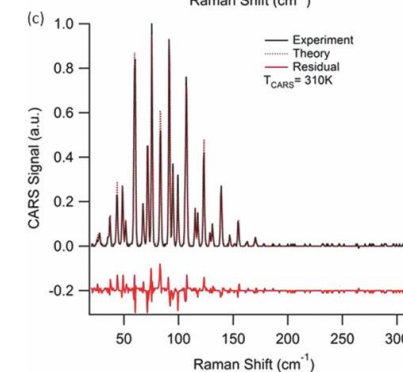
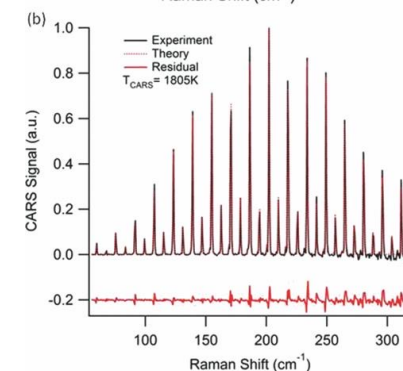
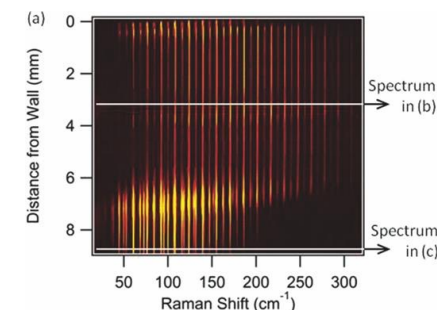
$$\frac{\Delta\nu}{\nu} = \frac{\Delta\lambda}{\lambda} = \frac{10^{12} \text{ (Hz)}}{3 \times 10^8 \text{ m/s} / (780 \times 10^{-6}) \text{ m/s}} \approx 10^{-2} \quad \Delta\lambda \approx 8 \mu\text{m}$$

Pump (fs) and Stokes (ps) can be **aligned**
(contain wide range of frequencies in one chirp)
Separated from signal by splitter

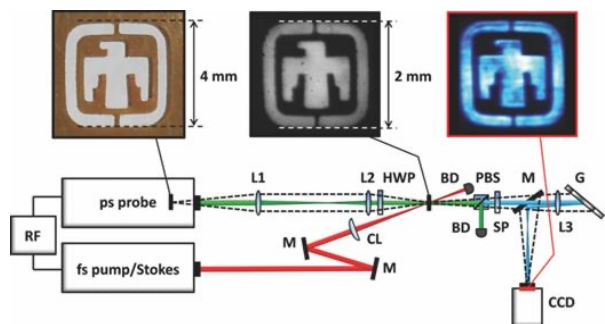


[Bohlin, A. Kliever, C. J. Chem. Phys. 138, 081102 \(2013\)](#)

1D CARS

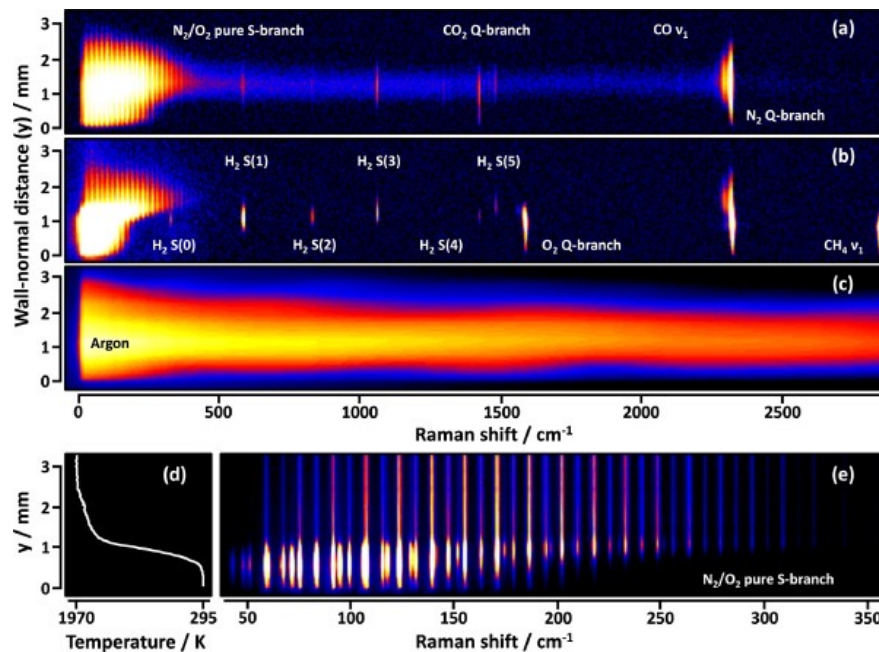


2D CARS

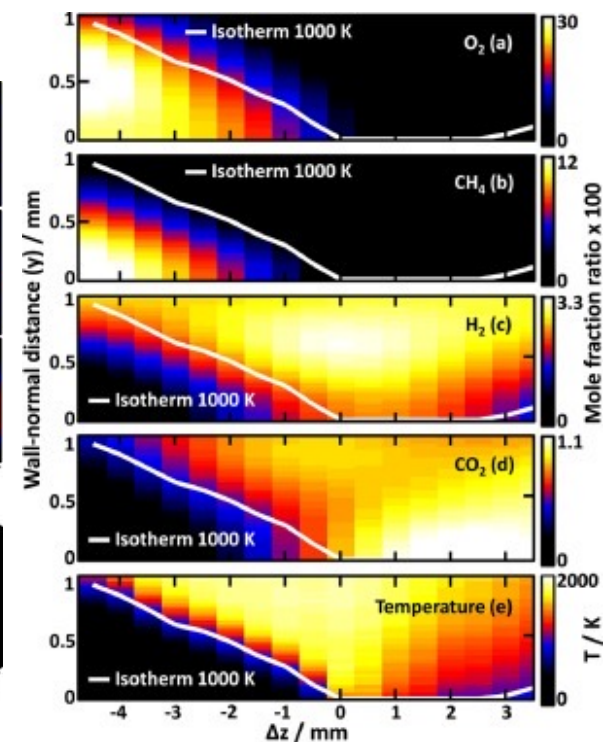


[Bohlin, A. and Klierer, C. J. Chem. Phys. 138, 221101 \(2013\)](#)

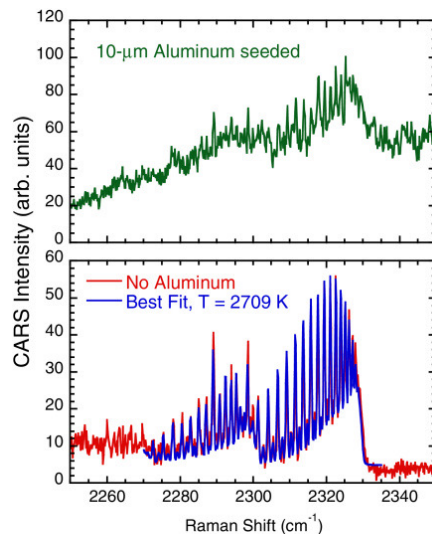
Temperature near walls



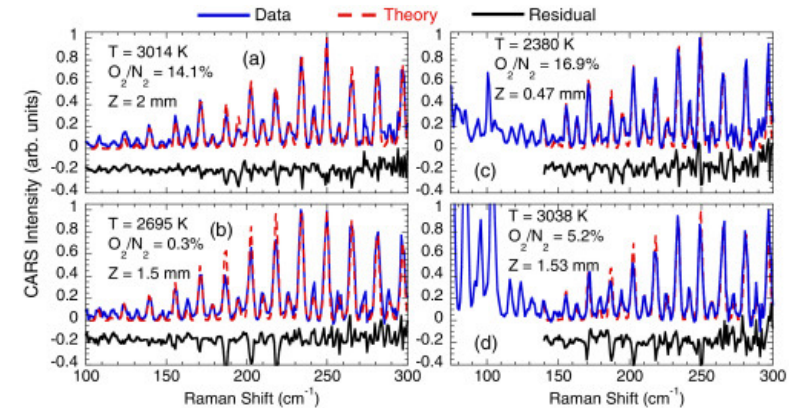
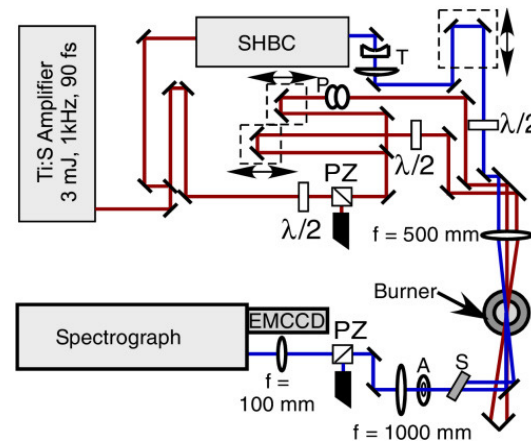
[Bohlin, A. et al., Proc. Comb. Inst. 36, 4557 \(2017\)](#)



CARS works in very noisy (luminous) environments



Nanosecond vibrational CARS spectra recorded from a C₂H₂/air flame



fs Vibrational CARS spectra
Probe delay removes resonant contribution

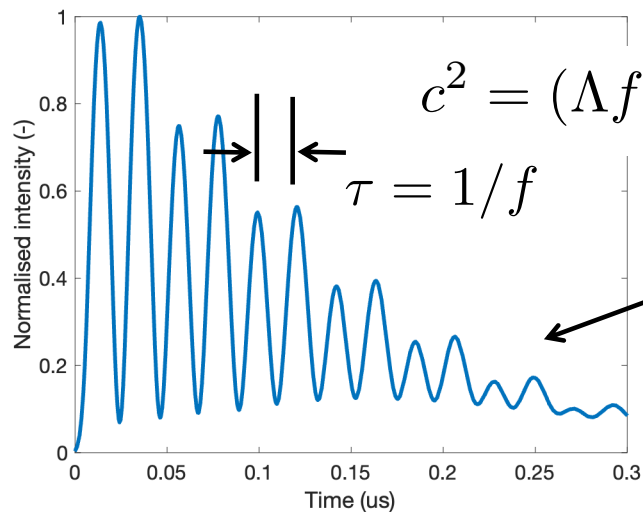
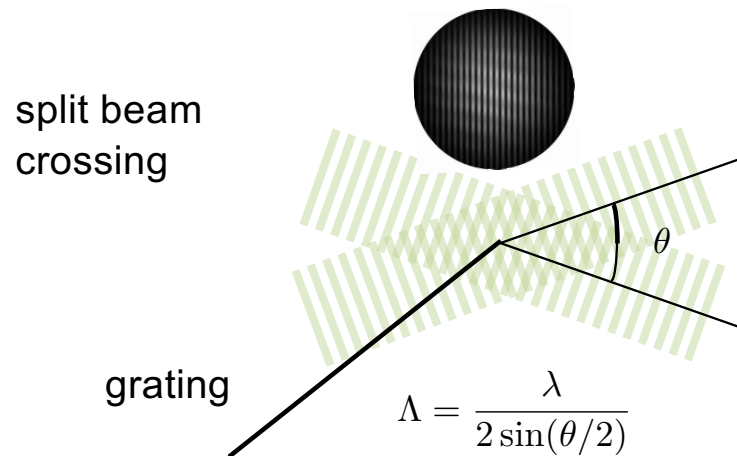
[Sean P. Kearney and Daniel R. Guildenbecher, "Temperature measurements in metalized propellant combustion using hybrid fs/ps coherent anti-Stokes Raman scattering," Appl. Opt. 55, 4958-4966 \(2016\)](#)

Fs-CARS: allows broadband detection of species and temperature
time delay removes resonances

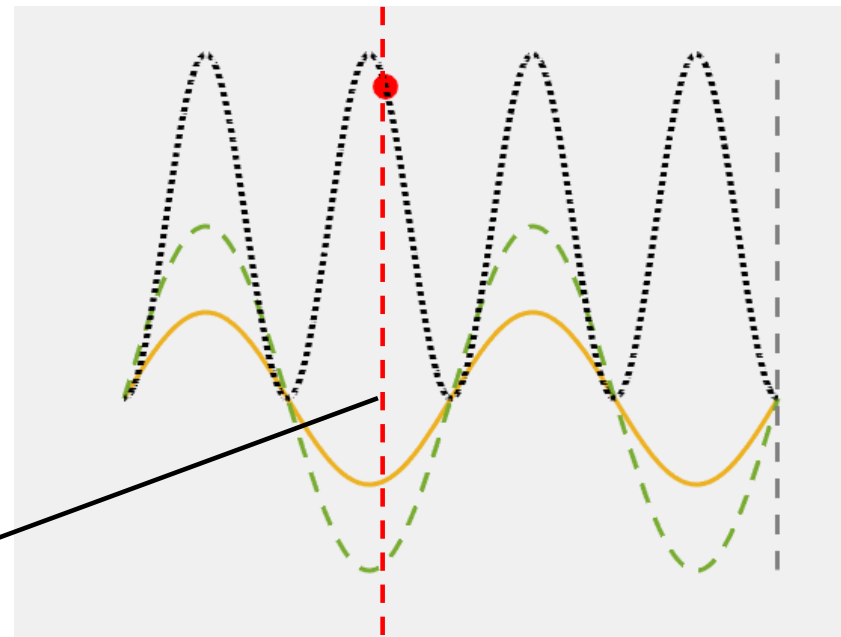
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Laser Induced Thermal Grating Spectroscopy



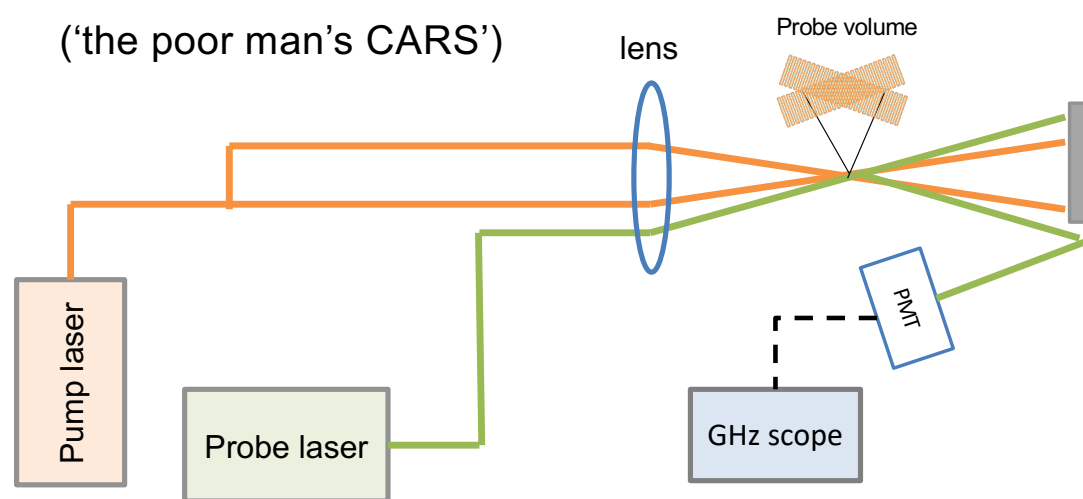
Density perturbation



stationary
moving at c
sum
 $\Delta \text{Density}^2$

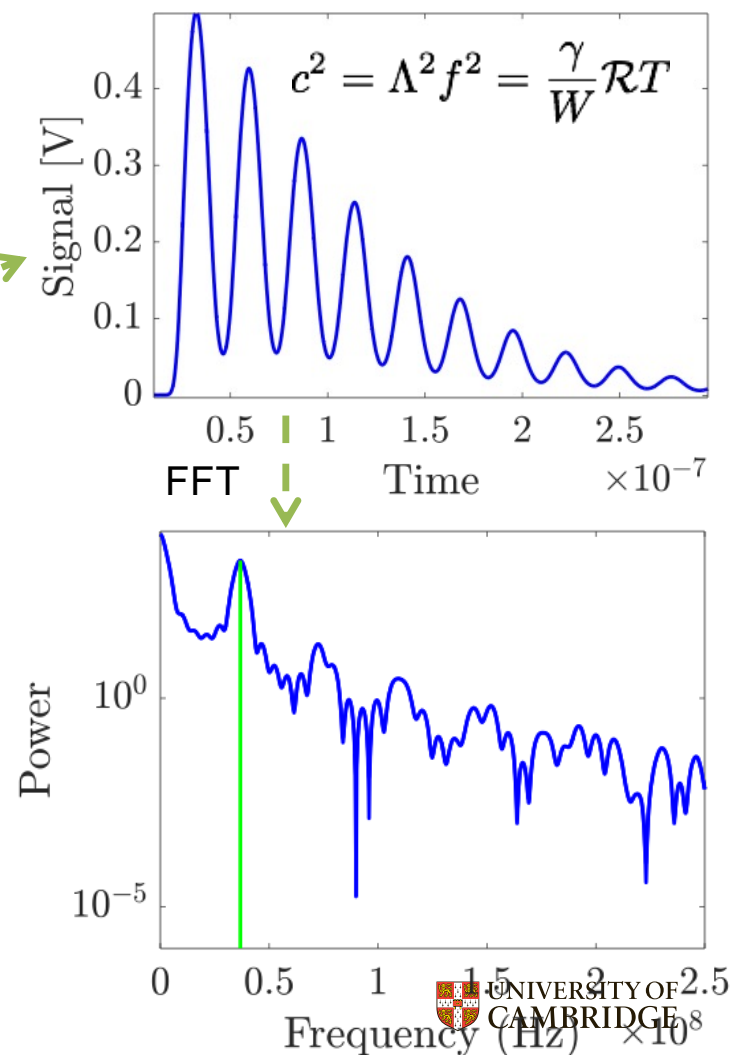
Frequencies can be measured very accurately
Temperature is accurate as γ/W .

Laser-induced grating spectroscopy for temperature measurements



Frequencies can be measured VERY accurately (10^{-6})
 Speed of sound is a standard for calibration of temperature
 Can be used in sooty environments!

Spatial resolution around 1-2 mm
 Requires high power and absorbing substance (H₂O ok)
 Best signal at pressures above 3 bar



Thermal (LITGS) and electrostrictive (LIEGS) behaviour

Thermal: strong signal produced by **local absorption by a gas**: when present, produces peaks at periods of

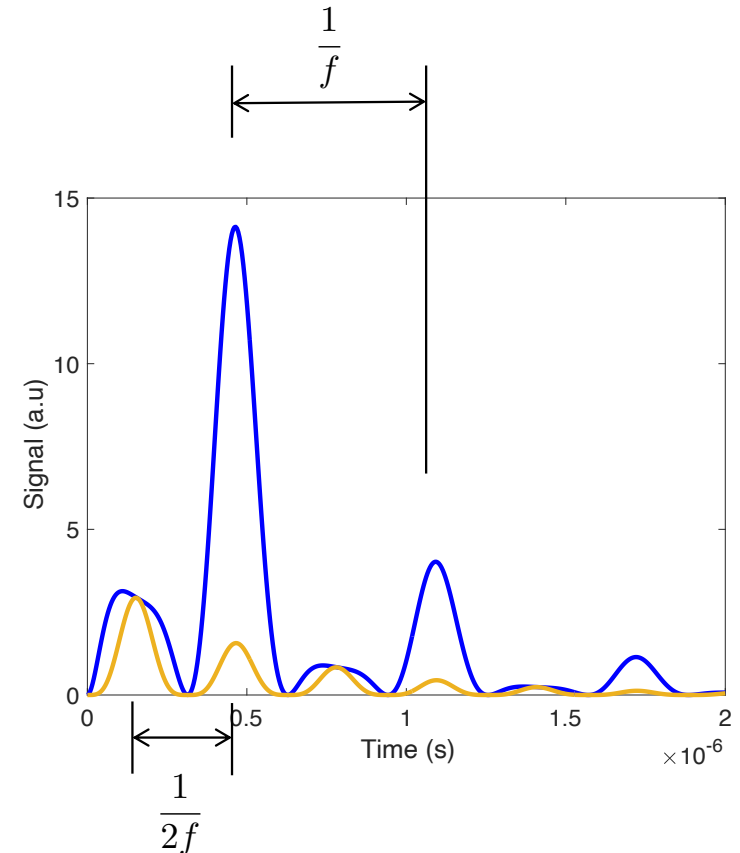
$$\tau = 1/f \quad c^2 = (f\Lambda)^2 = \frac{\gamma\mathcal{R}}{W}T$$

Electrostrictive: weaker signal produced by local change in density owing to **strong electric field**. Always present, but sometimes not quite detectable, at a harmonic

$$\tau = \frac{1}{2f} \quad c^2 = (2f\Lambda)^2 = \frac{\gamma\mathcal{R}}{W}T$$

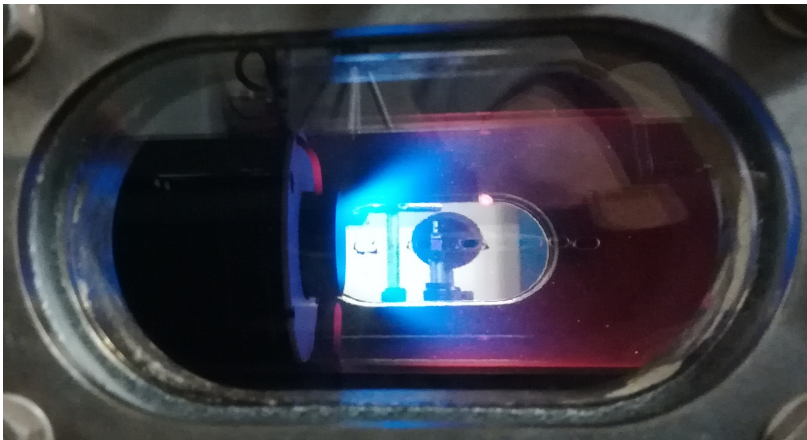
$$T = T_0 \left(\frac{n_0}{n} \frac{f}{f_0} \right) \frac{W/\gamma}{W_0/\gamma_0}$$

$n = 1$ thermal
 $n = 2$ electrostrictive



Combining **thermal** and **electrostrictive** it is possible to get temperatures in the reactants and products. Not yet suitable for very high spatial resolution, but very accurate.

Cardiff GTRC: Hydrogen/Methane Premixed

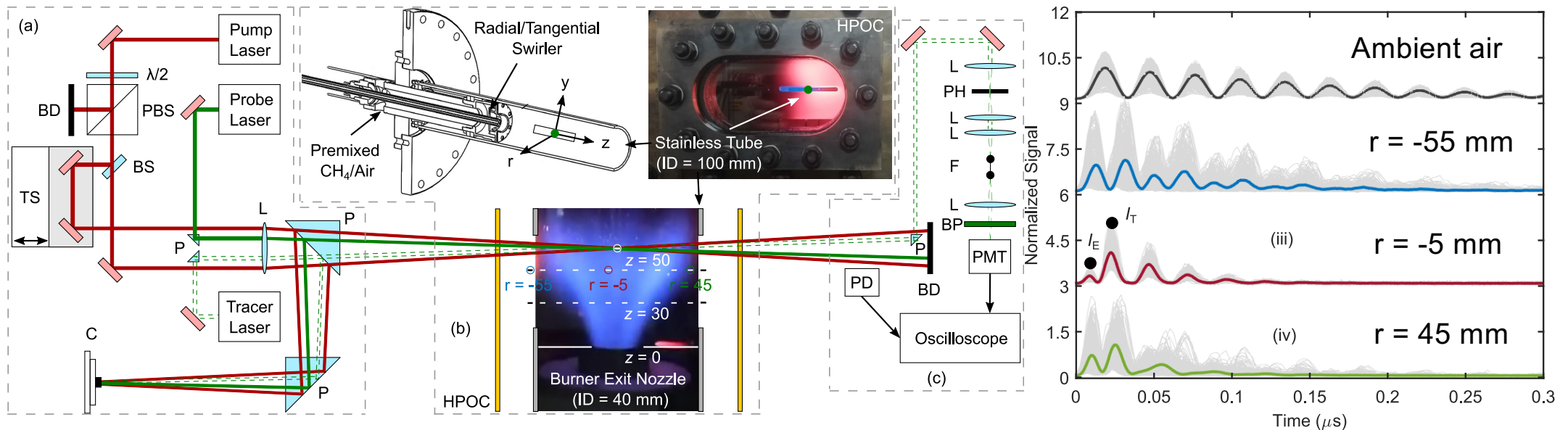


Hydrogen 60%
Methane 40%
 $P = 3 \text{ bar}$
 $T = 500 \text{ K}$



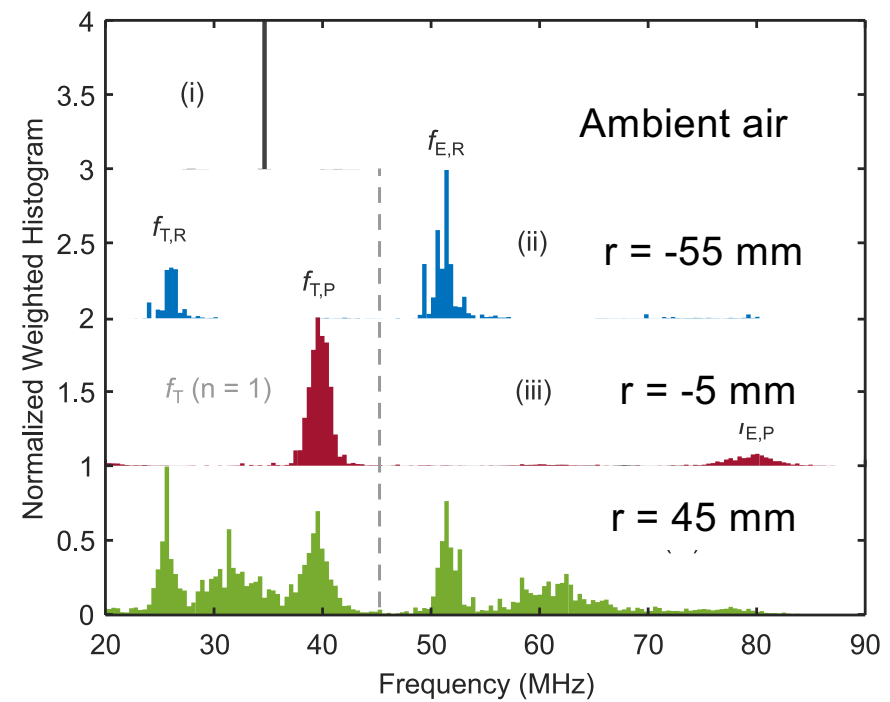
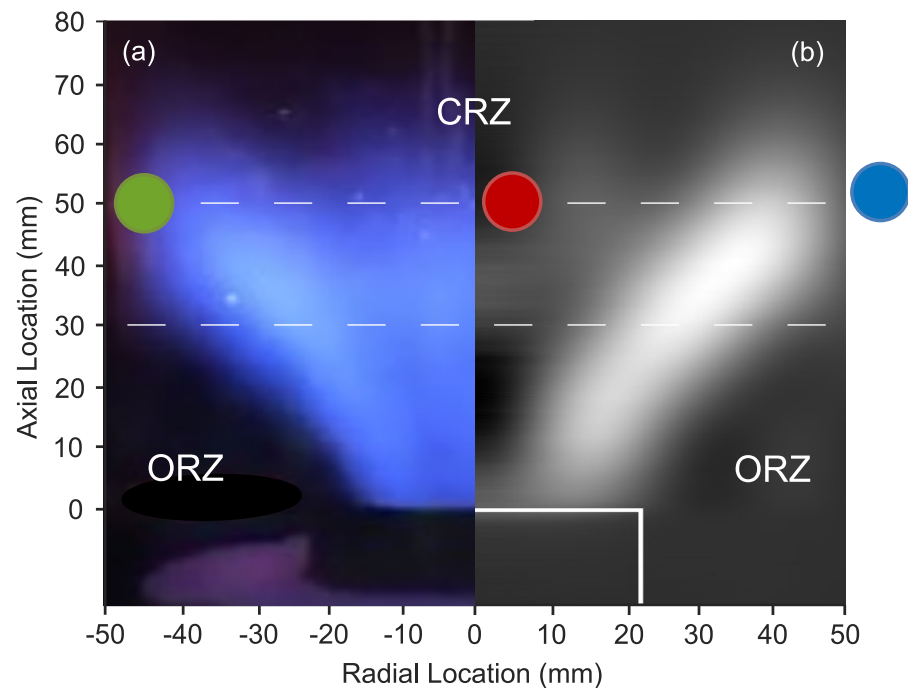
GTRC Port Talbot

LITGS applied to gas turbine combustor measurements



1064 nm pump (excites water)
 ~ 70 mJ/pump
 532 nm probe, ~ 7 W

LITGS in gas turbine combustor



Extracting temperatures and water vapour molar fractions from LITGS signals

$$T = T_0 \left(\frac{n_0}{n} \frac{f}{f_0} \right) \frac{W/\gamma}{W_0/\gamma_0}$$

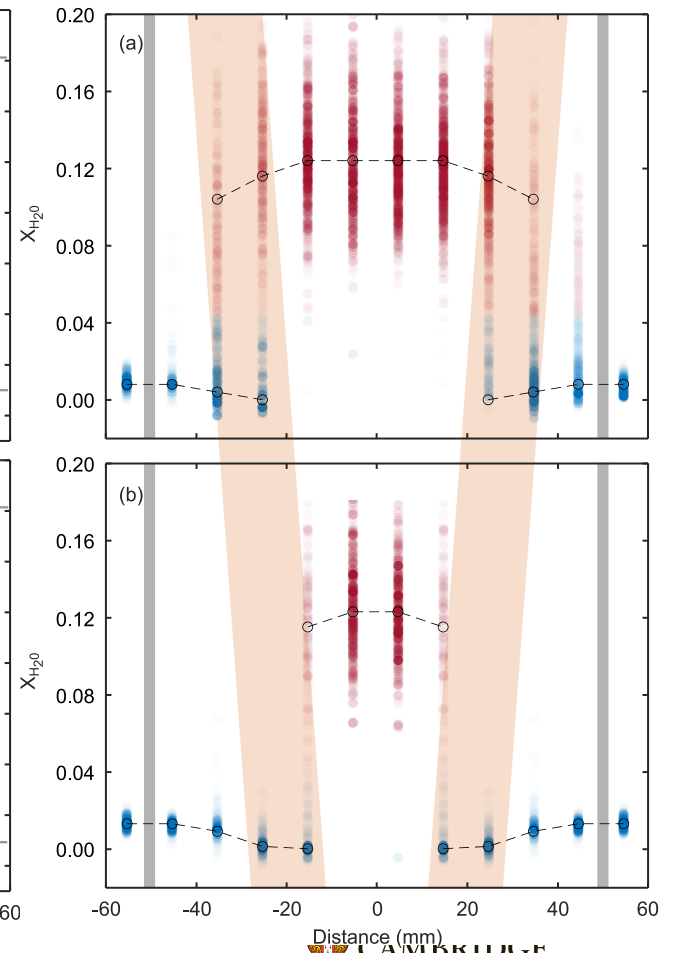
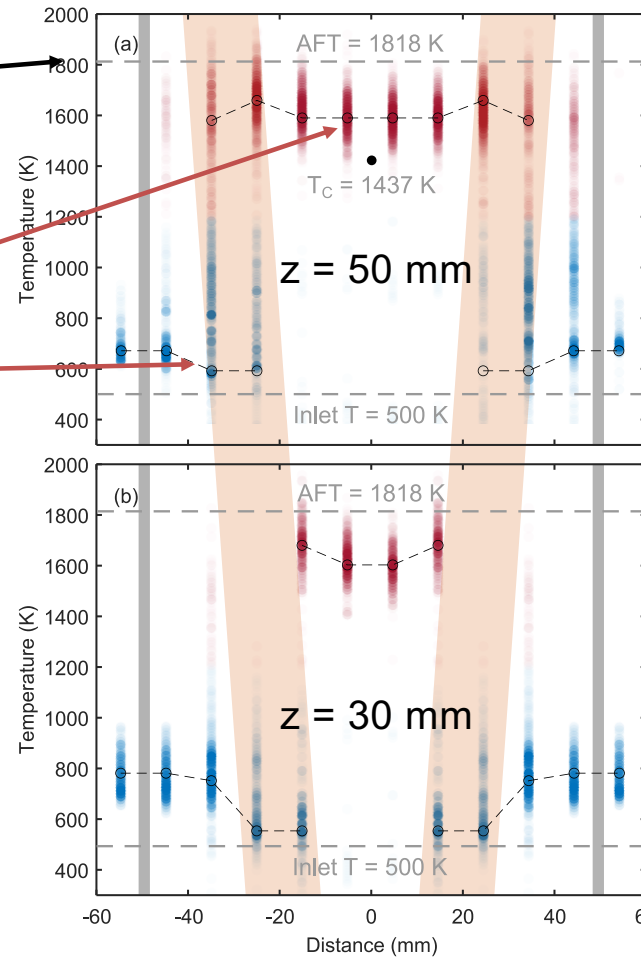
stoichiometric

thermal

electrostrictive

Temperature

Water vapour



Proceedings of ASME Turbo Expo 2023
Turbomachinery Technical Conference and Exposition
GT2023
June 26-30, 2023, Boston, Massachusetts

GT2023-101185

SPATIAL TEMPERATURE AND WATER MOLAR CONCENTRATION MEASUREMENTS USING THERMAL AND ELECTROSTRICTIVE LIGS DURING OPERATION OF A SWIRL BURNER AT PRESSURE

Lee Weller^{1,*}, Priyav Shah², Anthony Giles³, Francesca De Domenico⁴, Steve Morris², Benjamin A. O. Williams², Simone Hochgreb¹

¹Department of Engineering, University of Cambridge, Cambridge CB2 1PZ, UK

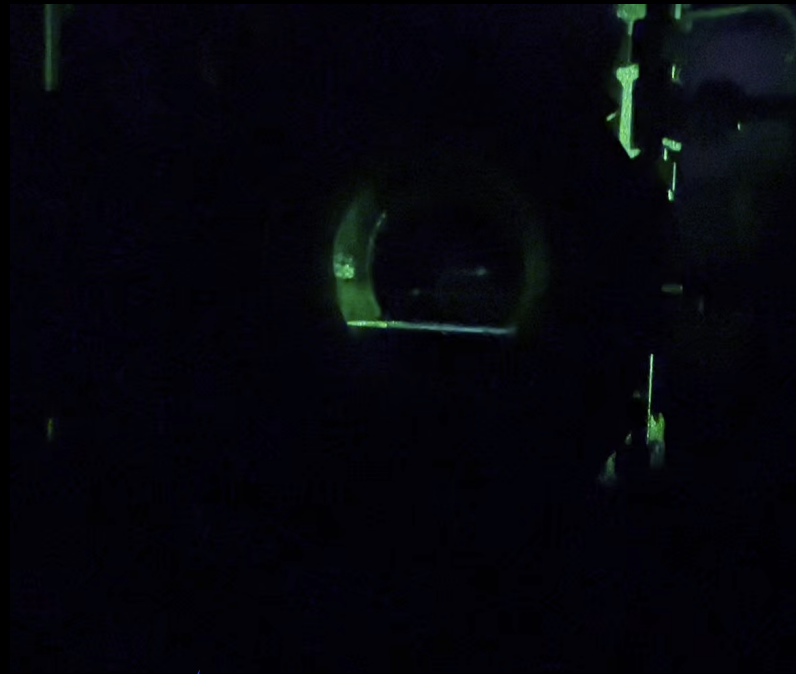
²Department of Engineering, University of Oxford, Oxford OX1 3PJ, UK

³School of Engineering, Cardiff University, Cardiff CF24 3AA, UK

⁴FPT, Aerospace Engineering, TU Delft, Delft, 2629HS, NL

CAMBRIDGE

Time-resolved measurements of combustion instabilities in sooty flames



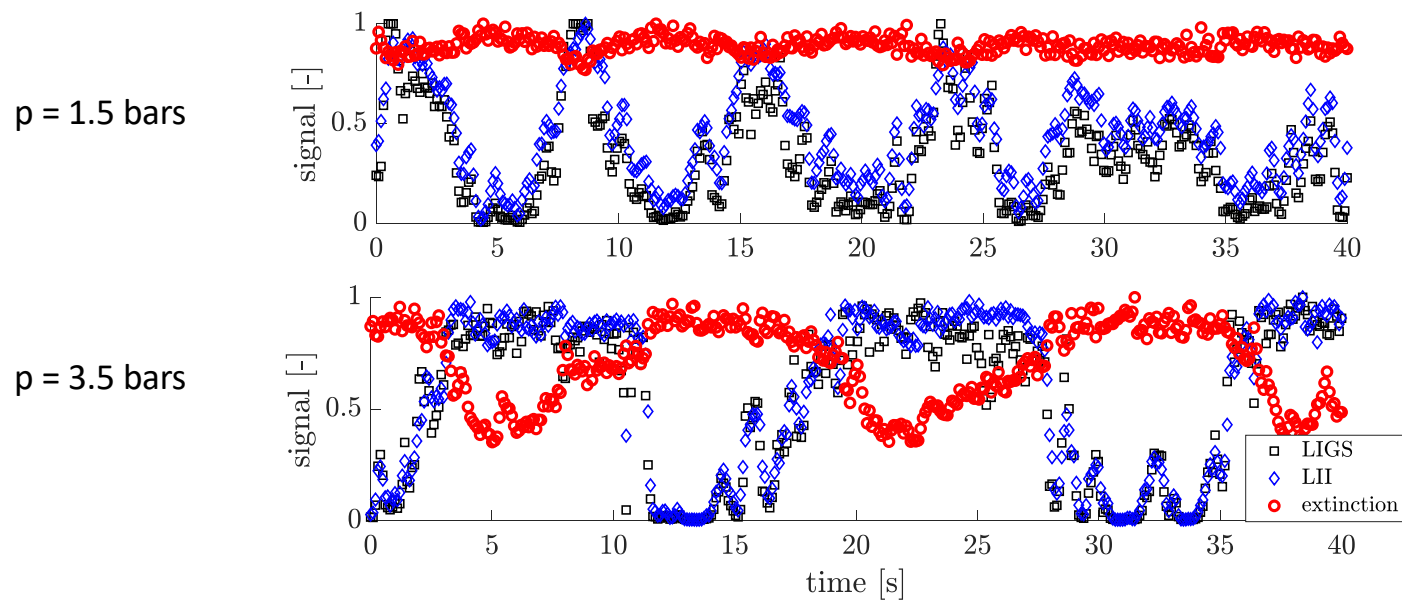
Methane/air
Ethylene/air
 $p = 4$ bar



Water absorbs 1064 nm light

Soot is a broad absorber as a black body

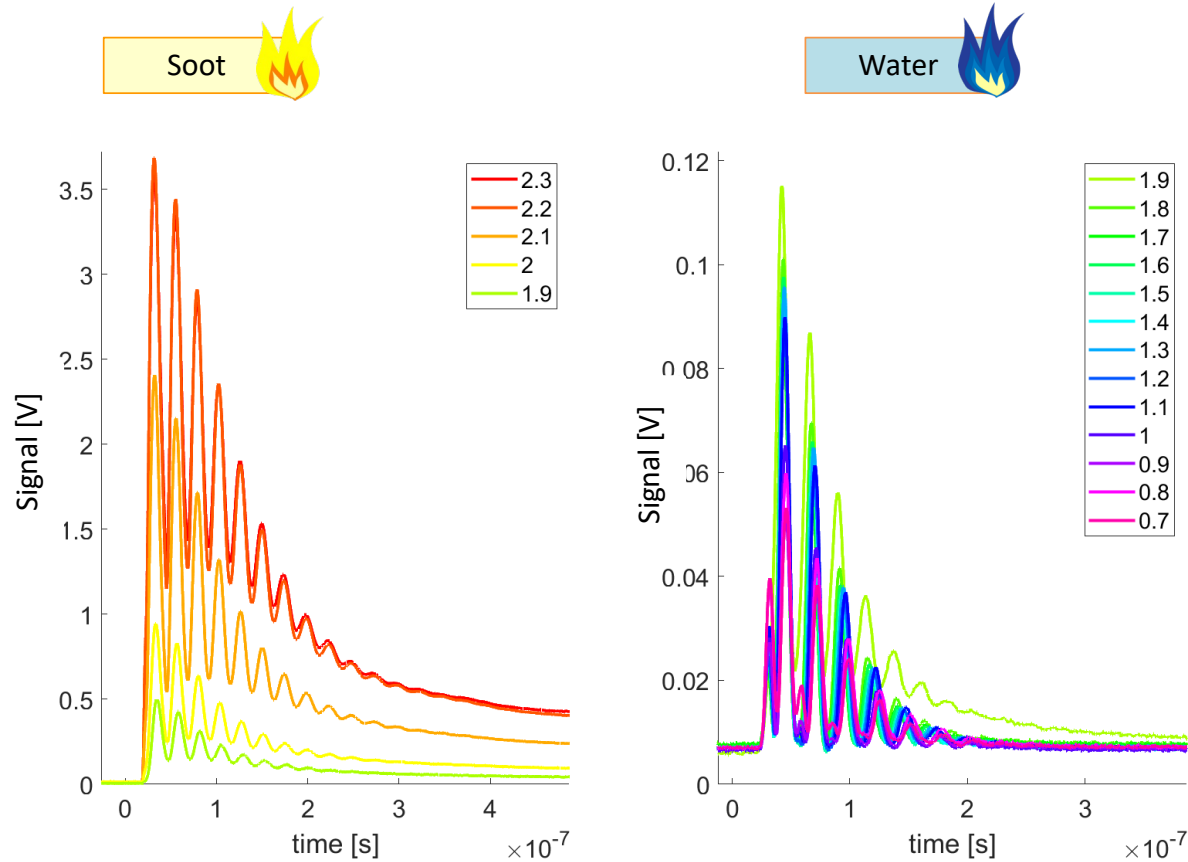
LITGS with soot and water vapour absorption: unsteady flames



Work in progress: can we extract further information from the probe volume to understand the local composition (H_2O , soot) as well as temperature?

LITGS equivalence ratio scan

SCAN of the FUEL MASS FLOW RATE while keeping the air mass flow rate and pressure constant, $p = 3$ bar



Courtesy:
Lee Weller,
Fran De Domenico

Questions



So is it always better to use coherent methods?



But what is the advantage of getting a coherent beam?



If fs/ps CARS seems so powerful, why not use it everywhere?



LITGS looks very useful, why have I not heard of it?

Useful Reviews

- Springer Handbook of Experimental Fluid Mechanics, Tropea, C., Yarin, A. L. and Foss, J. (Eds), Springer, 2007, <http://www.springer.com/materials/mechanics/book/978-3-540-25141-5>
- Westerweel, J., G.E. Elsinga, R.J. Adrian, Particle Image Velocimetry for Complex and Turbulent Flows, Annual Reviews of Fluid Mechanics (2013). <http://www.annualreviews.org/doi/full/10.1146/annurev-fluid-120710-101204>
- Roy, S., J.R. Gord, A.K. Patnaik, Recent advances in coherent anti-Stokes Raman scattering spectroscopy: Fundamental developments and applications in reacting flows, Prog. Energy Combust. Sci. 36 (2010) 280–306 doi:10.1016/j.pecs.2009.11.001. <http://www.sciencedirect.com/science/article/pii/S0360128509000586>
- Schulz, C., V. Sick, Tracer-LIF diagnostics: quantitative measurement of fuel concentration, temperature and fuel/air ratio in practical combustion systems, Prog. Energy Combust. Sci. 31 (2005) 75–121 doi:10.1016/j.pecs.2004.08.002. <http://linkinghub.elsevier.com/retrieve/pii/S0360128504000619>
- Schulz, C., B.F. Kock, M. Hofmann, H. Michelsen, S. Will, B. Bougie, et al., Laser-induced incandescence: recent trends and current questions, Appl. Phys. B. 83 (2006) 333–354 doi:10.1007/s00340-006-2260-8. <http://link.springer.com/10.1007/s00340-006-2260-8>.
- Frank, J. H., Advances in imaging of chemically reacting flows, J. Chem. Phys. 154, 040901 (2021), <https://doi.org/10.1063/5.0028249>.
- Goldenstein, C. S., Spearrin, R. M., Jeffries, J. B., Hanson, R. K. Infrared laser-absorption sensing for combustion gases, Progress in Energy and Combustion Science, 60 (2017) 132-176, <https://doi.org/10.1016/j.pecs.2016.12.002>

Summary



Effective use of measurement techniques requires:

Understanding physical principles

Attention to detail in the setup

Use most recent models for understanding uncertainties



Technique should be selected based on:

What is the question?

What is the accuracy required to answer the question?

Tradeoff between complexity and accuracy

Sometimes the more complex and expensive does not give the best answer

The End

