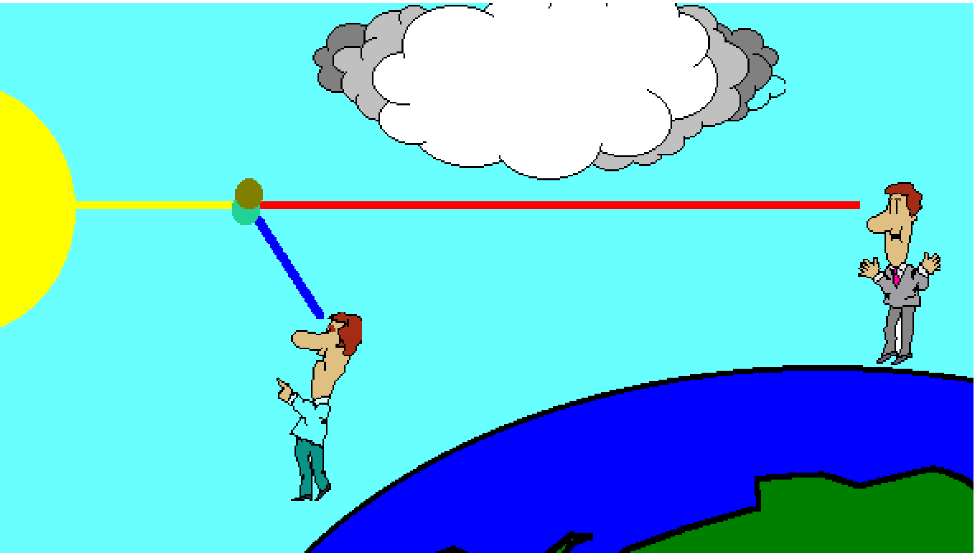
7. Rayleigh scattering

- Introduction
- Rayleigh scattering
  - Theory
  - Species concentration measurements
  - Temperature measurements
- Filtered Rayleigh scattering
- Summary

Introduction: Why is the sky blue?



- Daytime sky looks blue on a clear day
- The sky looks red at sunset/sunrise

Why?

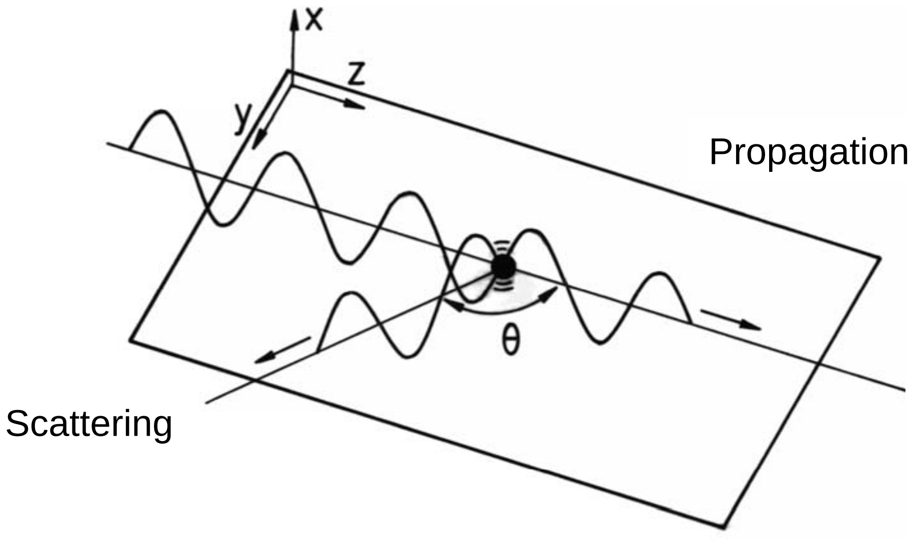
Rayleigh scattering!!

The scattering intensity is proportional to λ^{-4}

A wavelength at 430 nm (in the blue) is thus scattered a factor of ~6 times as efficient as a wavelength of 680 nm (in the red).

The physical principle

Electromagnetic wave propagating along the z-axis. The polarization is vertical (along x-axis). The scattering in the y-z-plane is vertically polarized and of equal intensity.



$\mu = \alpha E$

α; polarizability

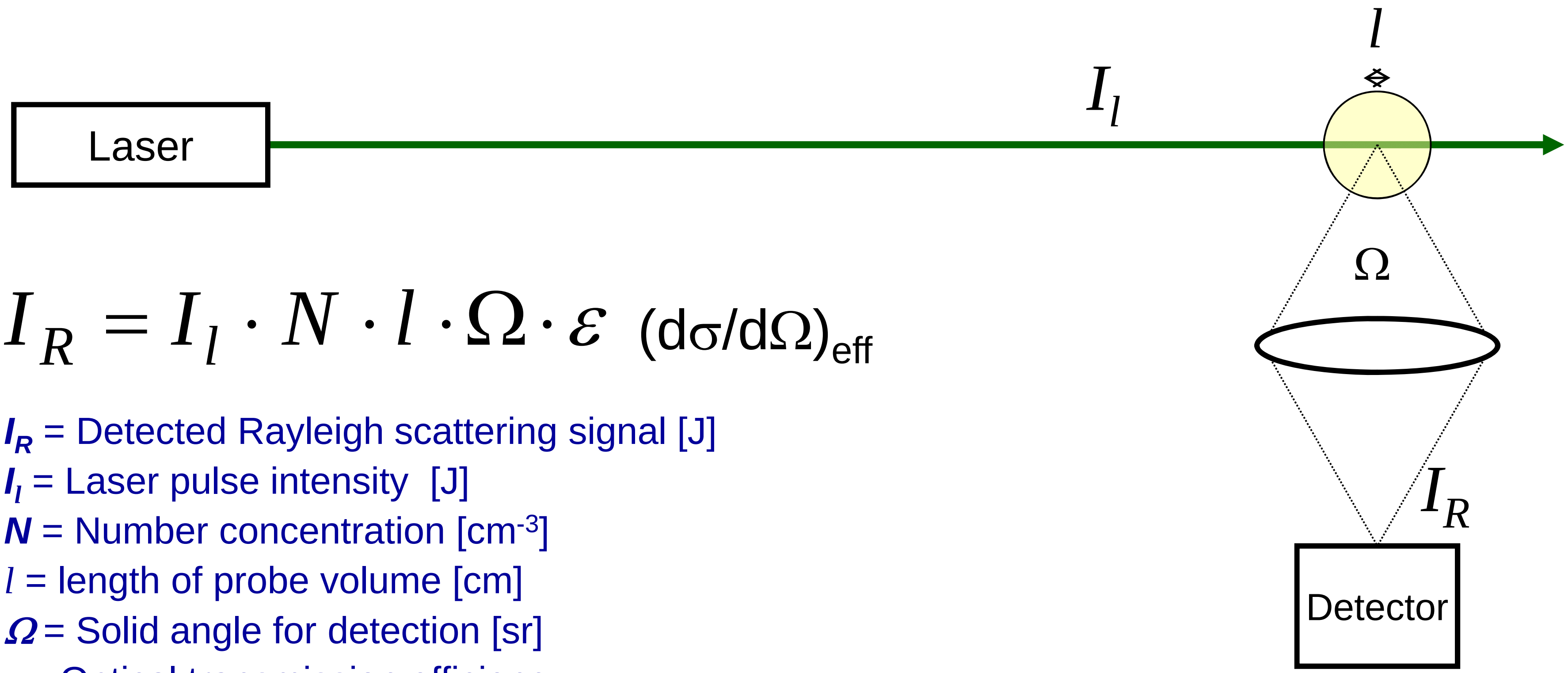
- **When an electromagnetic wave interacts with an atom/molecule/particle, the oscillating electric field creates an oscillating dipole, μ , when the electrons are moved back and forth.**
- **An oscillating dipole radiates at the same frequency as the incident radiation, $E = E_o \sin 2\pi\nu$, what is called Rayleigh scattering.**
- **Different molecules scatter with different efficiencies, since molecules have different numbers of electrons, which also are bound in different configurations.**

Diagnostic potential

- Rayleigh scattering is mostly used for temperature measurements. 2-D measurements can be performed. Examples will be shown.

- The possibility to make concentration measurements is in general limited. The reason is that all molecules scatter at the same wavelength. However, when a species with very large cross-section (fuel) is probed, species visualization is possible

A Rayleigh scattering setup



$$I_R = I_l \cdot N \cdot l \cdot \Omega \cdot \varepsilon \; (d\sigma/d\Omega)_{eff}$$

I_R = Detected Rayleigh scattering signal [J]
I_l = Laser pulse intensity [J]
N = Number concentration [cm^{-3}]
l = length of probe volume [cm]
Ω = Solid angle for detection [sr]
ε = Optical transmission efficiency

$(d\sigma/d\Omega)_{eff}$ = Rayleigh cross section for gas mixture [cm^2/sr]

$$(d\sigma/d\Omega)_{eff} = \sum_i X_i \, (d\sigma_i/d\Omega)$$

X_i = Mole fraction of species *i*
σ_i = Rayleigh cross section of species *i* [cm^2/sr]

Rayleigh scattering thermometry (1)

$$I_R = I_l \cdot N \cdot l \cdot \Omega \cdot \varepsilon \cdot (d\sigma/d\Omega)_{eff}$$

The Rayleigh scattering signal is proportional to the number concentration of species and the cross section of the gas mixture.

If the cross section $(d\sigma/d\Omega)_{eff}$ is assumed to be constant: $\boxed{I \sim N}$

According to the perfect gas law: $N = \dfrac{p\,A_o}{R\,T}$

Since A_0 and R are constants, and pressure can be considered to be constant in a combustion situation, it means that:

Rayleigh scattering signal is inversely proportional to the temperature, i.e. $I_R \propto 1/T$

Rayleigh scattering thermometry (2)

$$I_R \propto 1/T$$

This expression can now be applied to a two-dimensional image of Rayleigh scattering.

Example

Assume an imaging Rayleigh measurement where
1) the temperature is 300 K in measurement point A.
2) the signal is a factor of five stronger in A than in B.

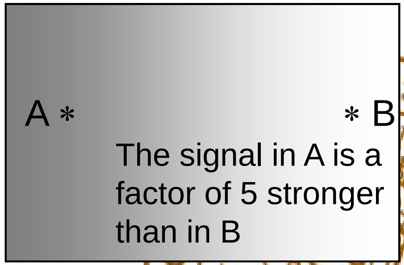
Real situation

A ∗ ∗ B
T_A= 300 K T = ?

Then we can calculate the temperature in point B:

$$T_B = T_A \frac{I_{R,A}}{I_{R,B}} = 300\ K \frac{5}{1} = 1500\ K$$

Imaged Rayleigh signal



The signal in A is a factor of 5 stronger than in B

Warning: Differences in Rayleigh cross sections for different species may give large errors!

Differential Rayleigh cross sections

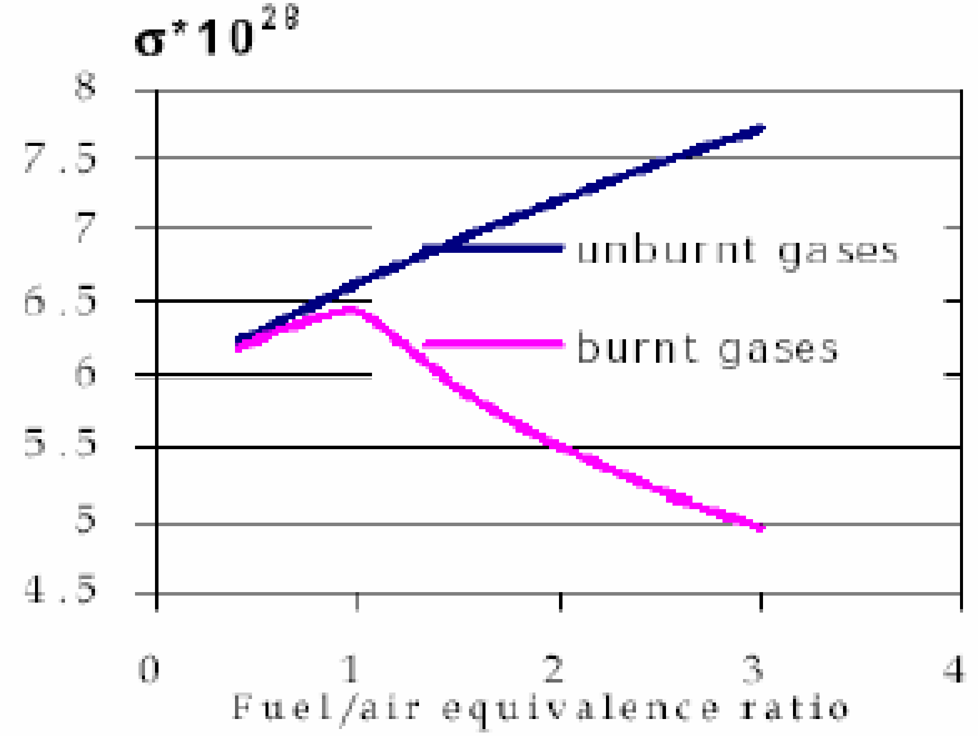
Differential Rayleigh cross sections for some different gases (for θ=90º at λ=532 nm, T=273 K, and p=1.013·10^5 Pa):

$$\left(\frac{\partial\sigma}{\partial\Omega}\right)_j = \frac{4\pi^2(n_j-1)^2}{N_0{}^2\lambda^4}$$

O_2	5.08 •10^{-28} cm^2/sr
N_2	6.13 •10^{-28} cm^2/sr
H_2	1.34 •10^{-28} cm^2/sr
CO	7.87 •10^{-28} cm^2/sr
CO_2	13.8 •10^{-28} cm^2/sr
H_2O	4.43 •10^{-28} cm^2/sr
C_3H_8	79.8 •10^{-28} cm^2/sr

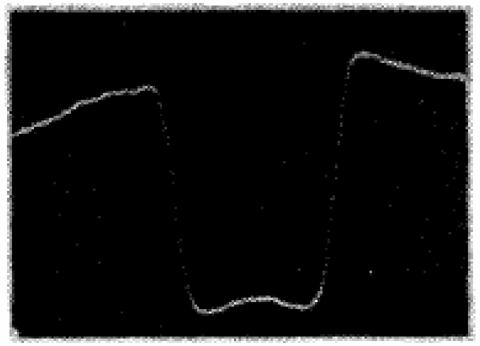
How can we measure in a flame with unknown composition?

Total Rayleigh scattering cross section in a methane/air flame

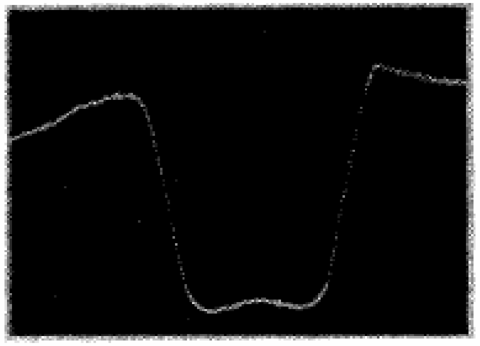


Spatially resolved Rayleich measurements along a line

Spatial Sensitivity
(No Flame)



h=10 mm



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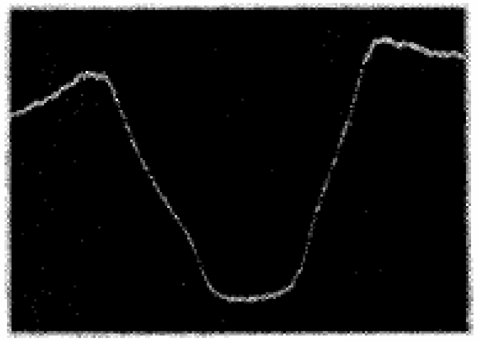
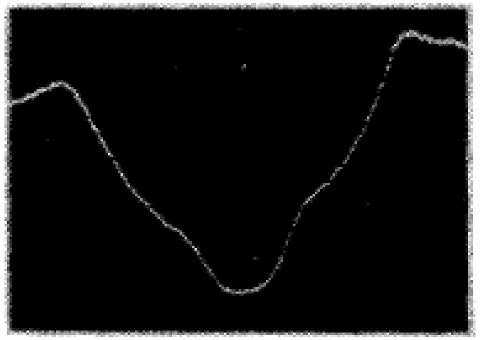


h= 40 mm



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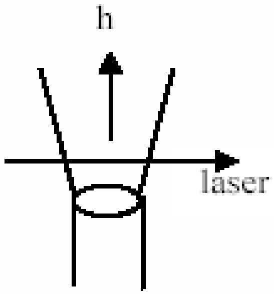
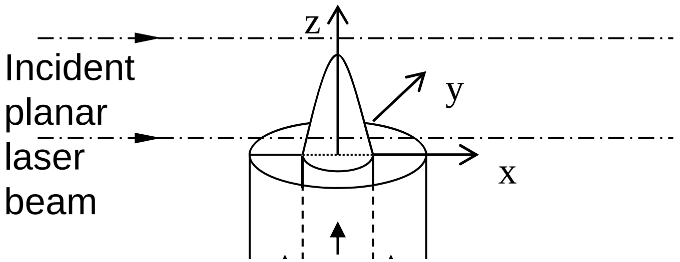
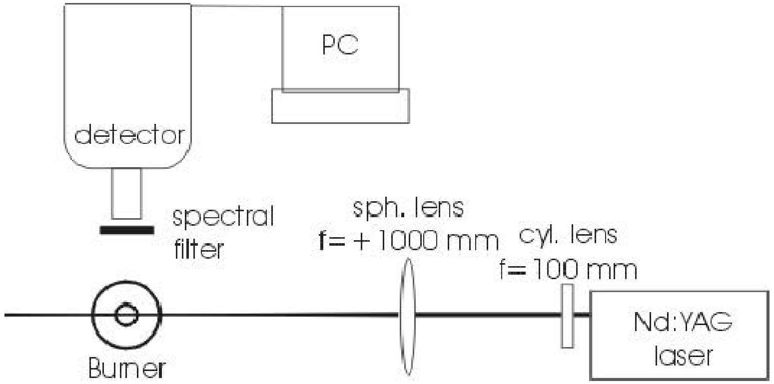
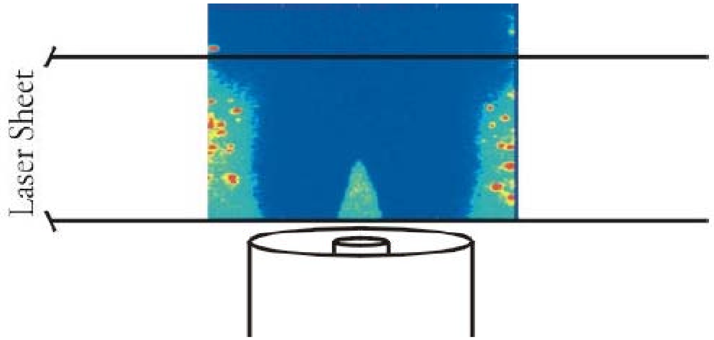
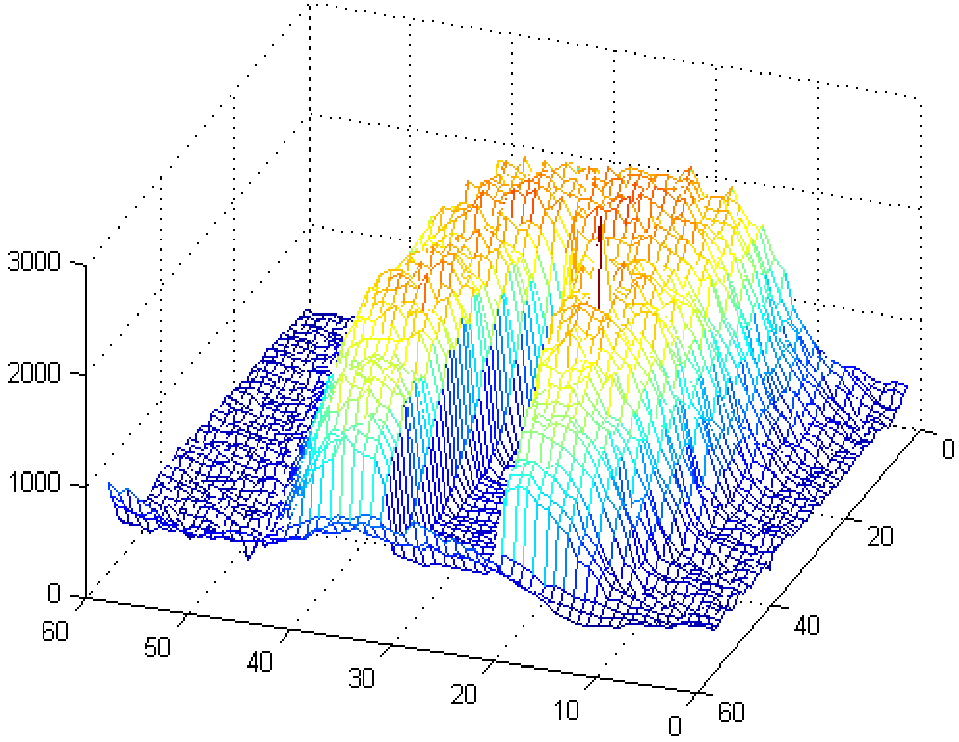


Example of 2D Rayleigh thermometry



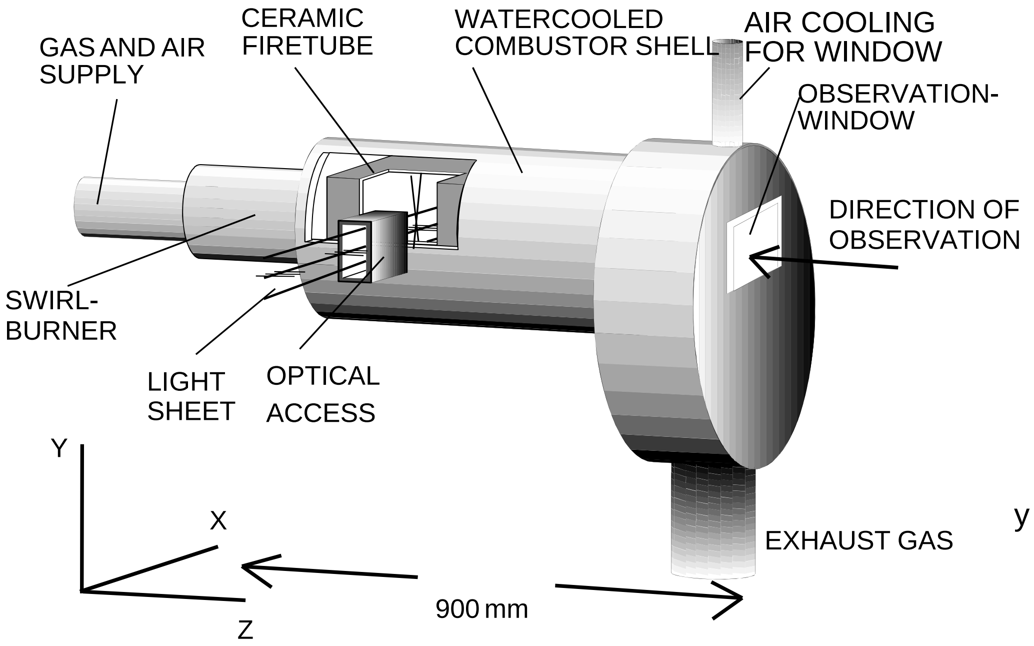


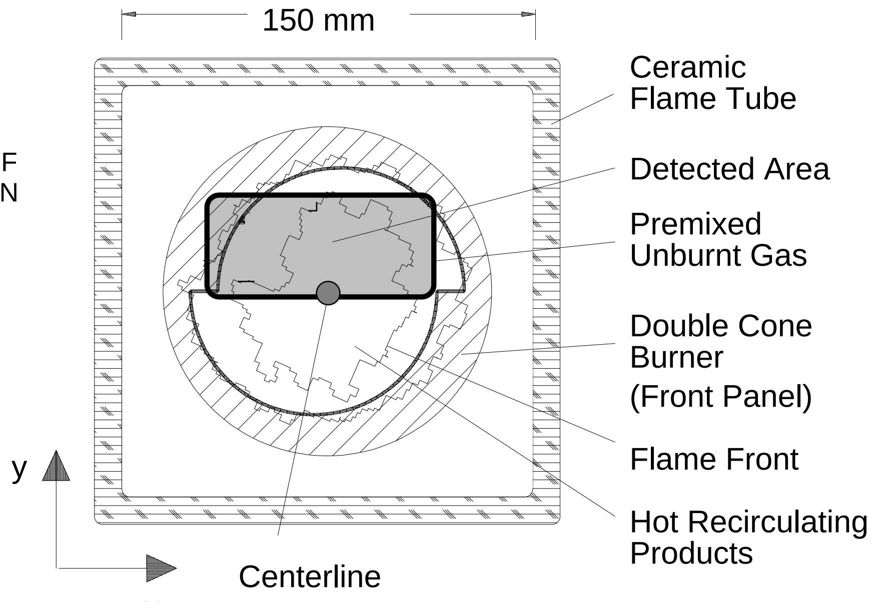




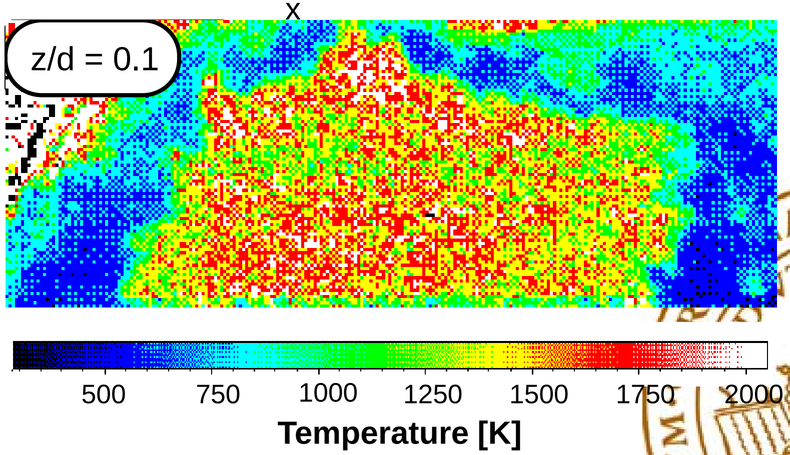
Final 2-D temperature plot

Rayleigh thermometry in combustor





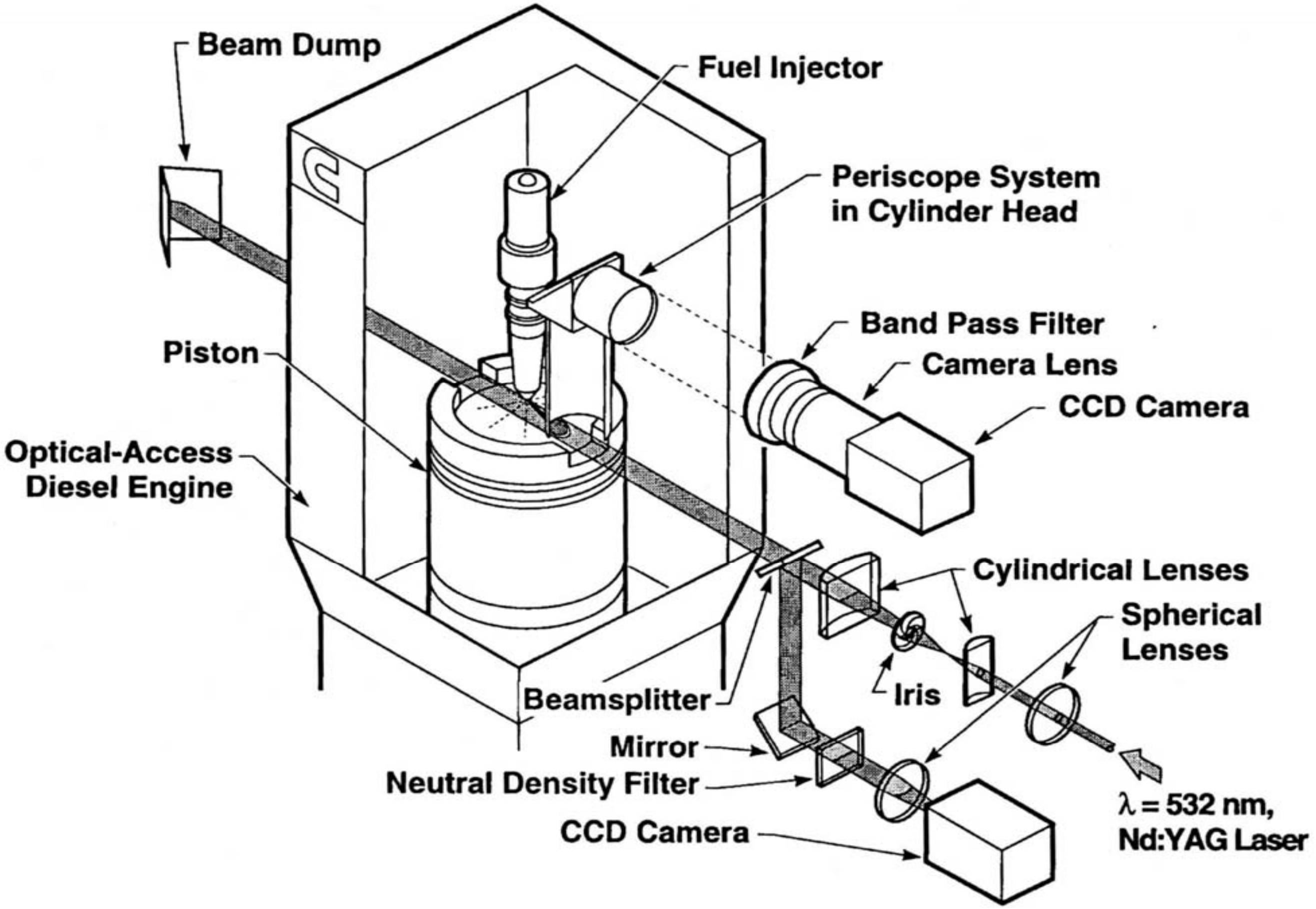
S. Kampmann, T. Seeger, and A. Leipertz, Appl. Opt. 34, 2780-2786 (1995)



Fuel concentration measurements

Under specific circumstances Rayleigh scattering can be used for concentration measurements.

Such a case is when there are few species with a big difference in Rayleigh cross section.

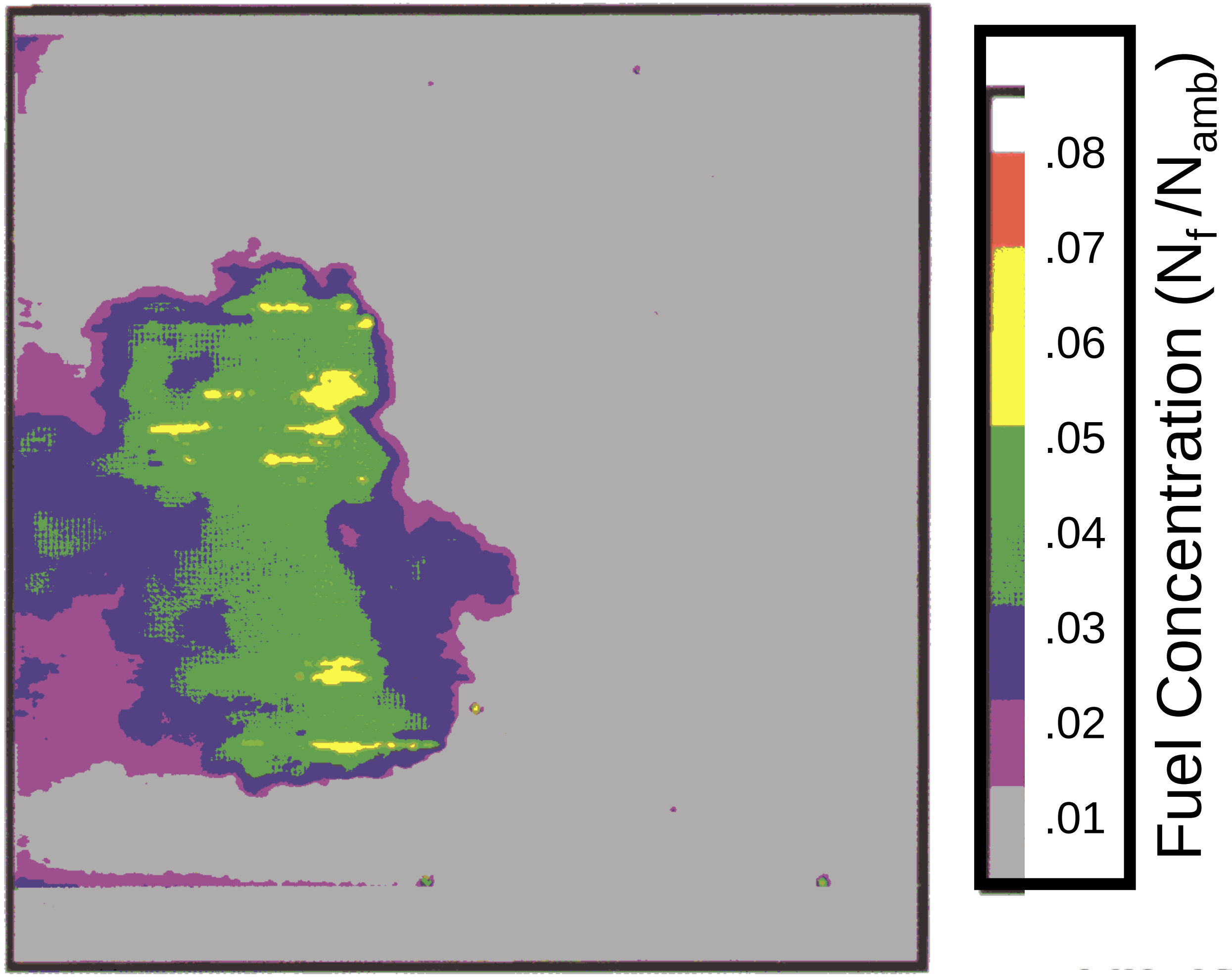


C. Espey, J. E. Dec, T.A. Litzinger, D.A. Santavicca, Combustion and Flame 109: 65-86 (1997).

Fuel concentration measurement in an engine

Big hydrocarbon molecules have much bigger Rayleigh scattering cross sections in comparison with small molecules such as N_2.

The ratio in Rayleigh cross section between a diesel fuel and air can be a factor of 300.

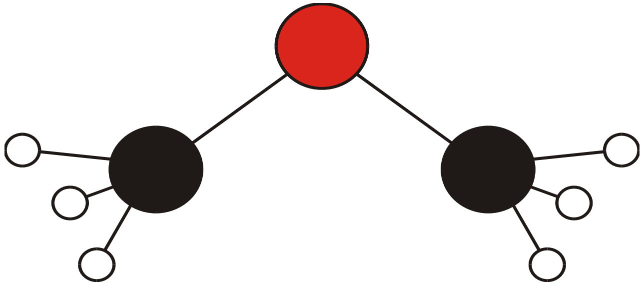


C. Espey, J. E. Dec, T.A. Litzinger, D.A. Santavicca, Combustion and Flame 109: 65-86 (1997).

DME spray imaging

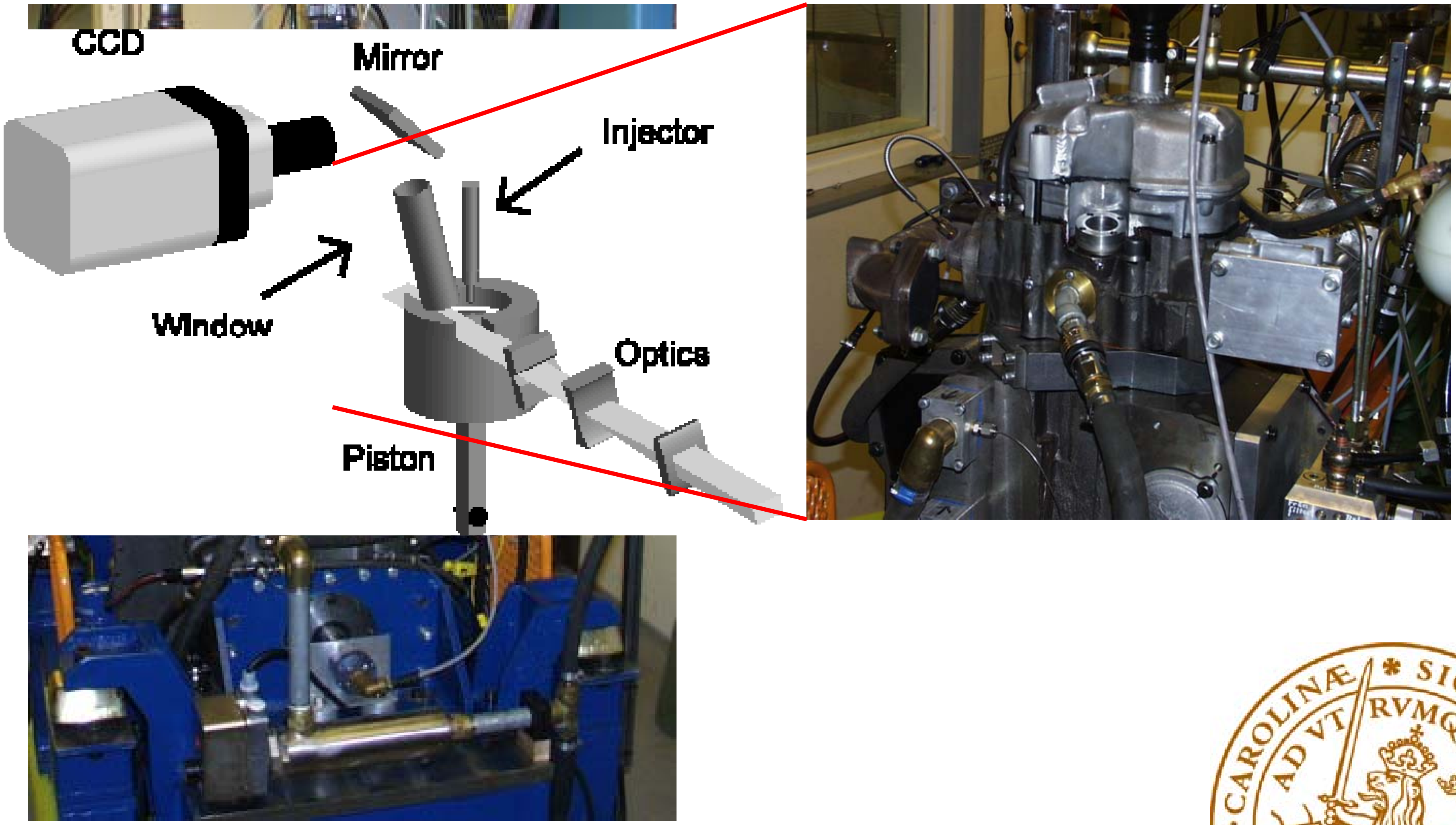
Dimethyl ether (DME) as alternative to diesel fuel:

- Good auto ignition characteristics
- Virtually eliminates soot
- Can substantially reduce NOx
- Can be produced in large quantities

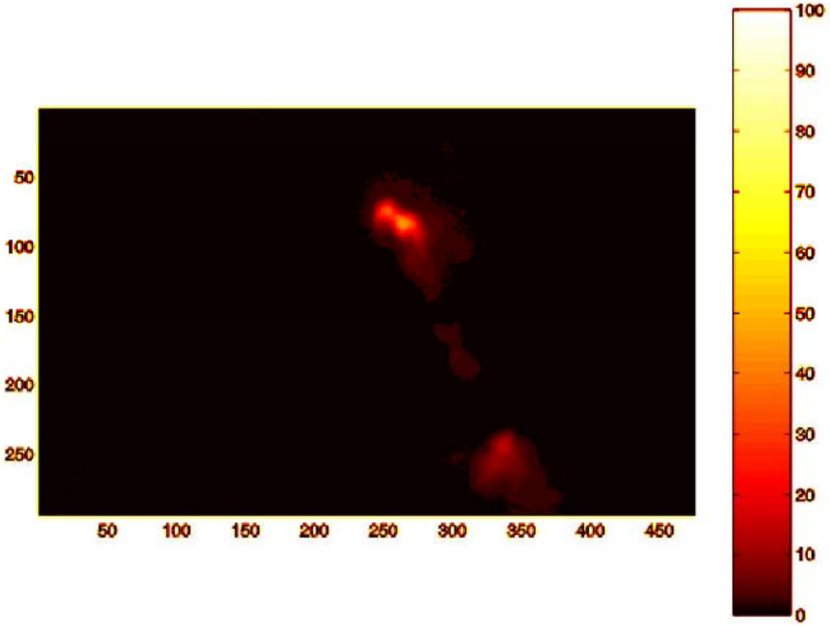


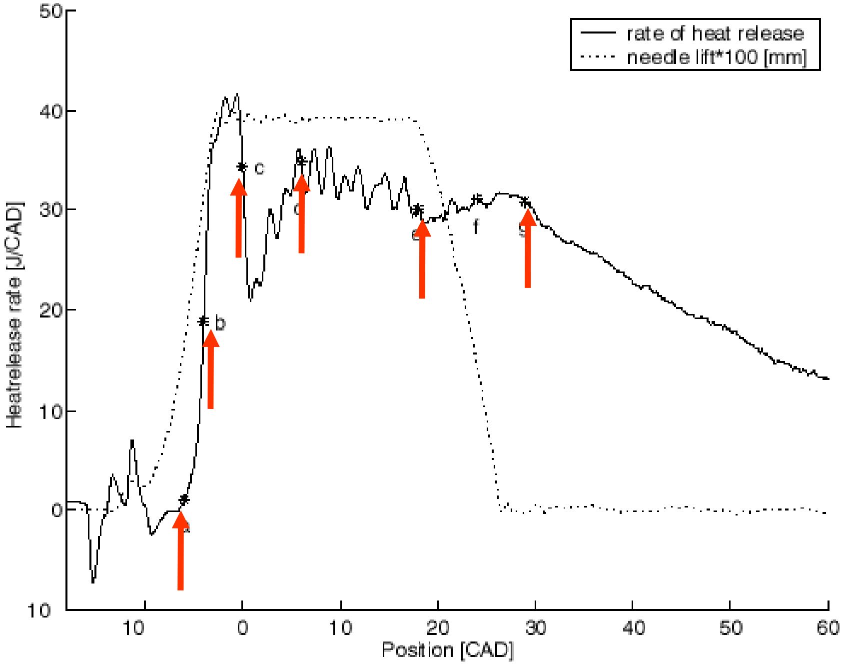
Aim: To investigate the mixing and ignition of DME

DME sprays in DI Diesel engine



Single-shot images from different engine cycles





Relative fuel concentrations

Rayleigh scattering: advantages

- It is an easy technique
- Arbitrary laser wavelength can be used, but shorter wavelengths leads to stronger signal (the λ^{-4}-dependence).
- Signal is proportional to number concentration → N and/or 1/T
- Signal is proportional to laser pulse energy, i.e. no quenching or saturation effects.

Limitations

- The technique is not species selective, since all atoms/molecules/particles scatter at the same wavelength.
- For accurate thermometry, the Rayleigh cross sections for individual species must be taken into account, which is hard work in a two-dimensional image since the mole fraction distribution must be known in every point.
- It is an incoherent technique
- Stray light from particles, optics and surfaces can interfere with the Rayleigh signal

Filtered Rayleigh scattering (FRS)

- The main problem with Rayleigh scattering is that scattering from optics, surfaces, particles, droplets interfere with the scattered light from molecules.

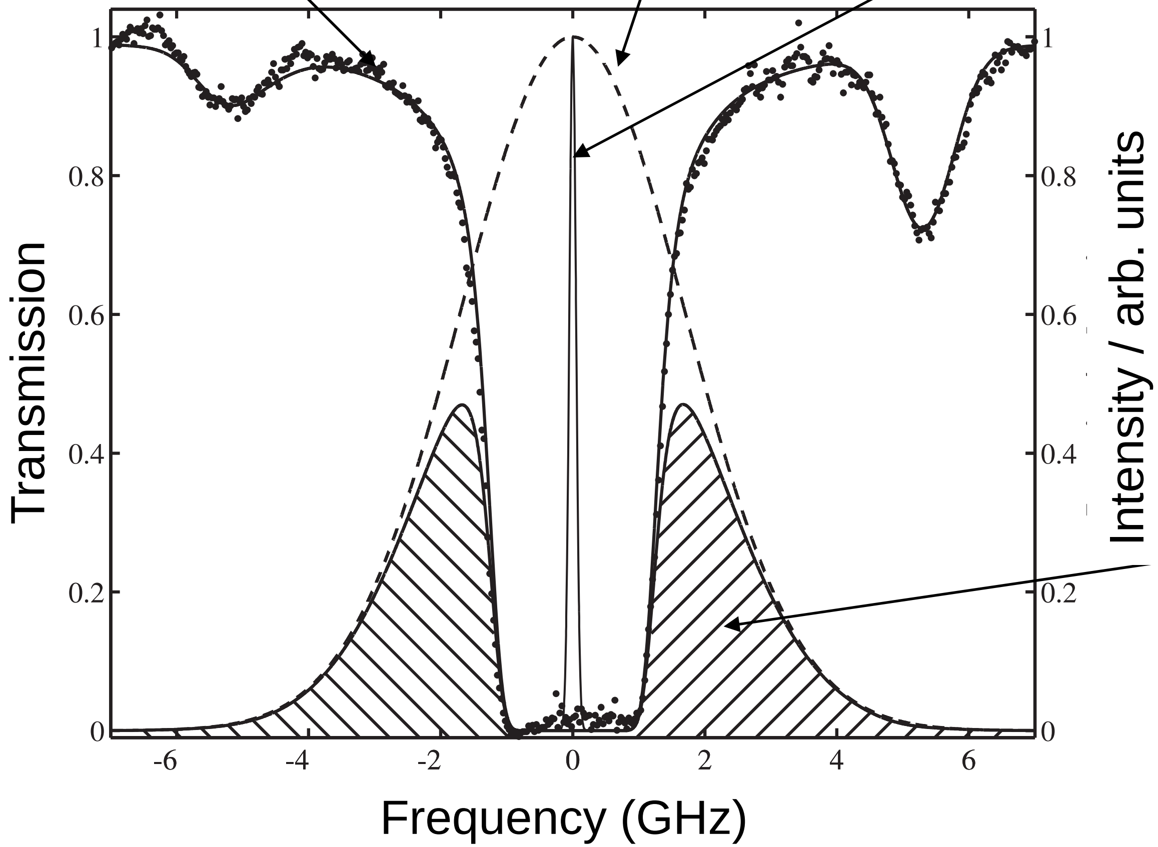
- This can be solved by the use of Filtered Rayleigh Scattering.

The principle for FRS

Transmission curve of a filter (mercury)

Rayleigh scattering line shape of N_2 at 1 atm. and 500 K

Scattering from particles and surfaces



Transmitted molecular Rayleigh scattering.

With increasing temperature, the line broadens and relatively more light passes the molecular filter.

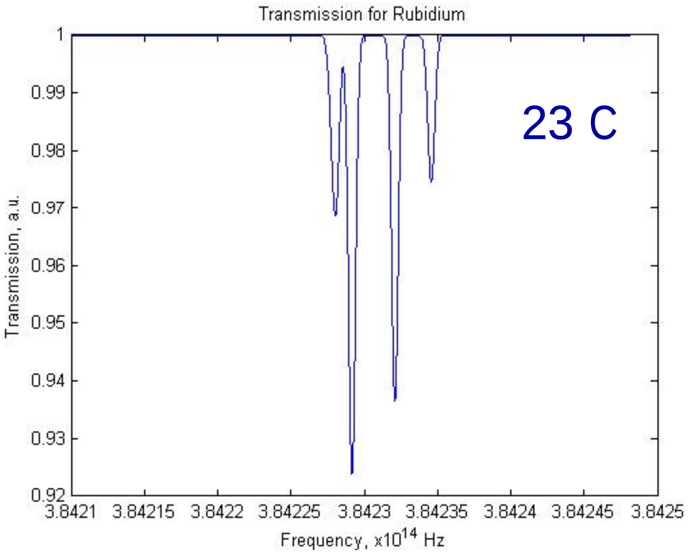
Possible filter candidates for FRS

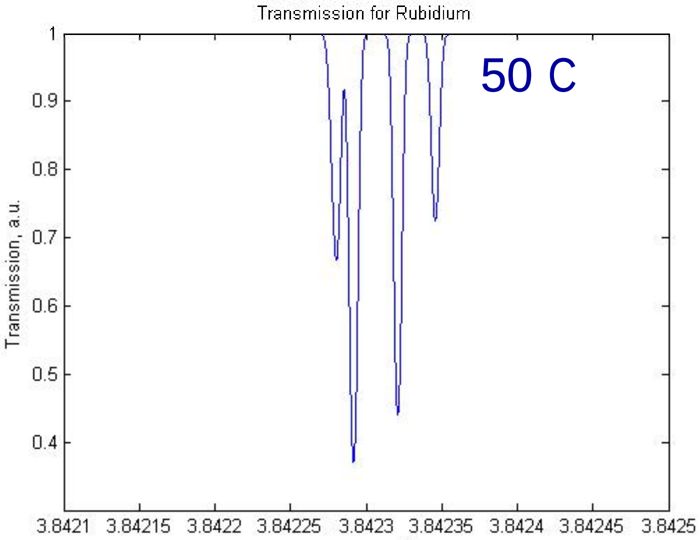
- **Mercury at 254 nm - Tripled Alexandrite laser**
  (+strong absorbing gas, +strong cross-section, +single isotope filter, +no spectroscopic fine structures, - medium laser energy, - molecular absorption, - ”exotic” laser)

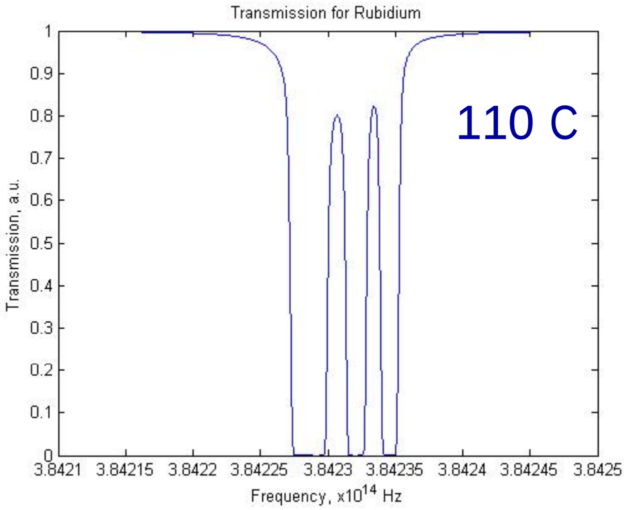
- **Iodine molecules at 532 nm – Doubled Nd:YAG**
  (+ easily available laser, +high laser energy, - medium absorbing gas, - medium cross-section, - rotational finestructures)

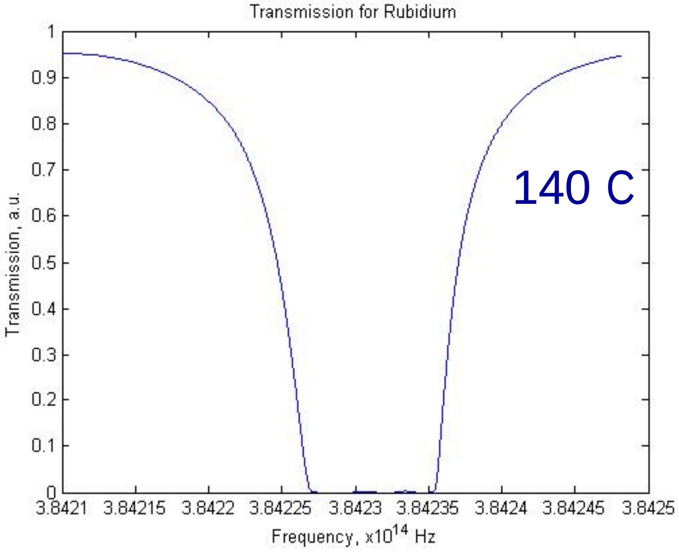
- **Rubidium 780 nm – Fundamental Alexandrite**
  (+strong absorbing gas, +single isotope filter, + no molecular absorption, + high laser energy, - spectroscopic fine structures, -weaker cross-section, - ”exotic” laser, - weak slope)

Transmission profiles at different T for Rubidium

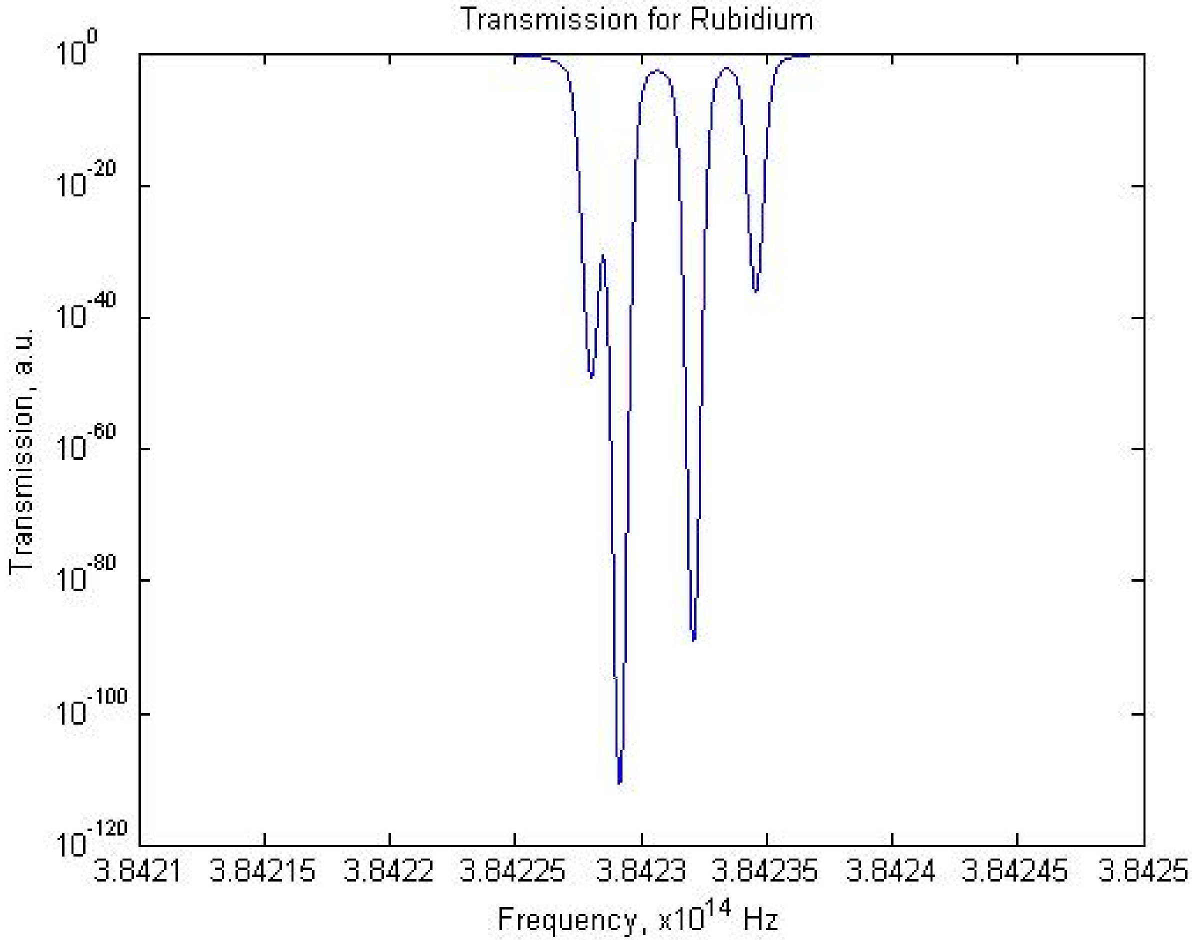




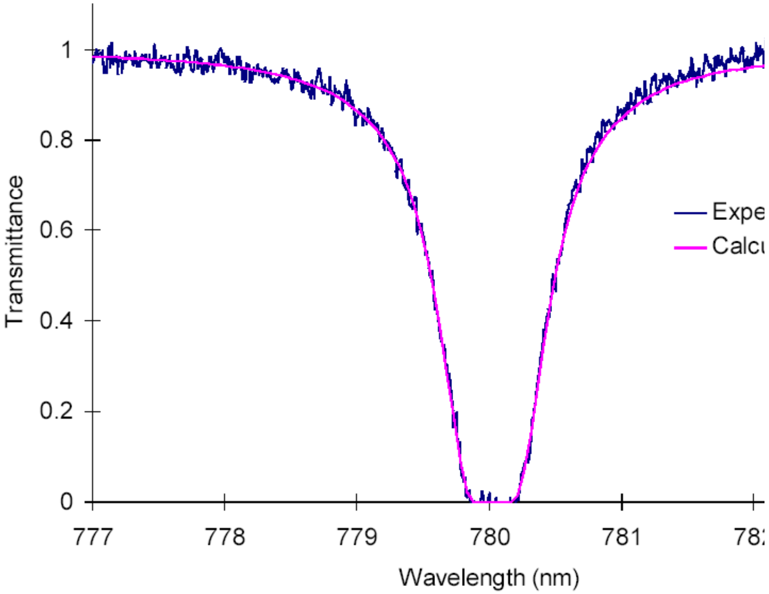


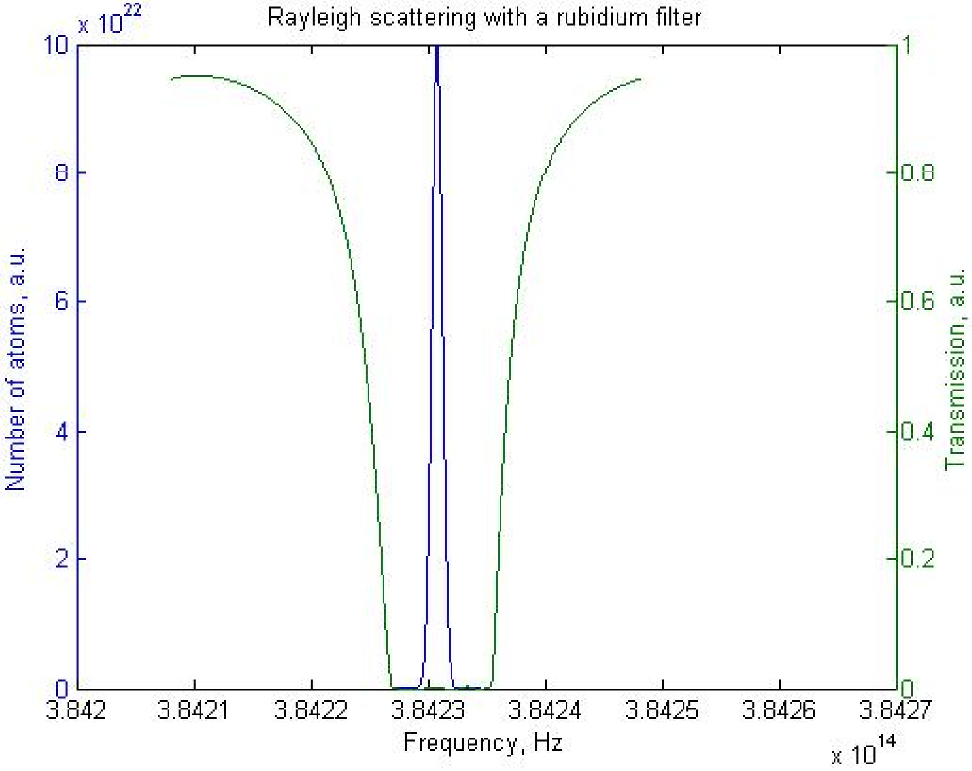


Transmission profile at 140 C (log scale)



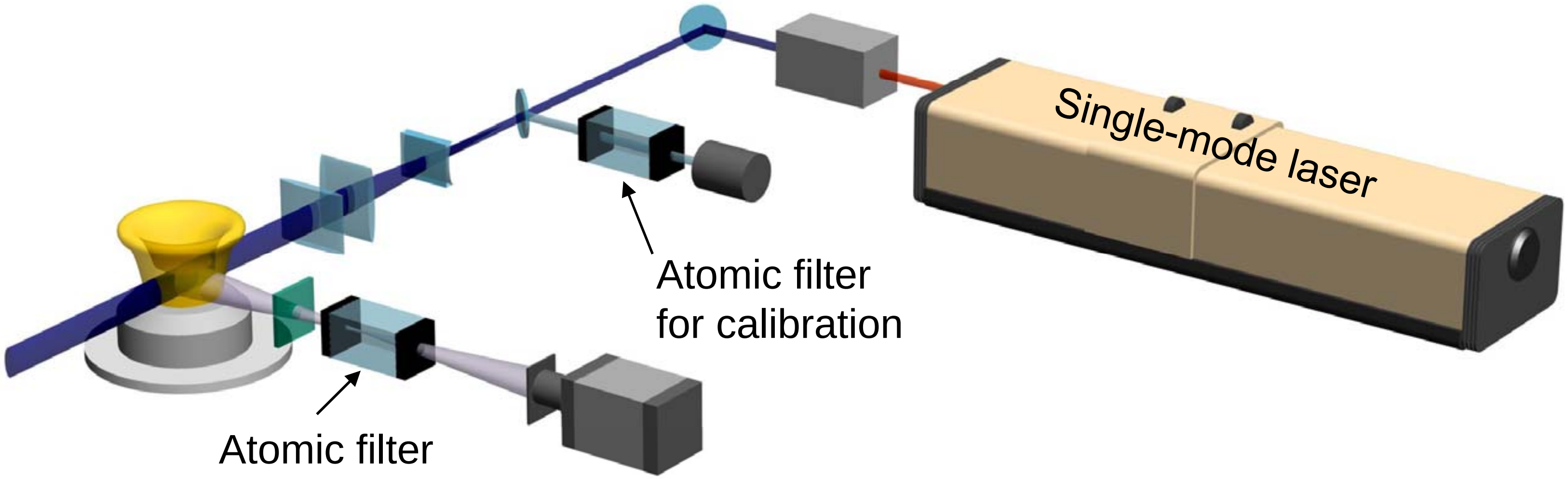
Limited use for FRS?





Experimental setup for FRS

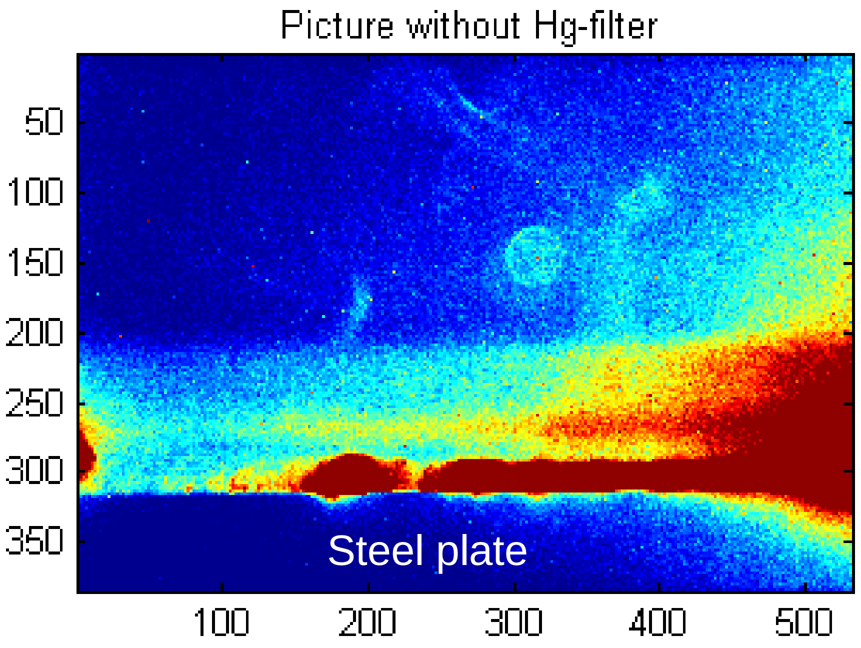
A filtered Rayleigh scattering setup is more complex than a normal Rayleigh scattering setup.

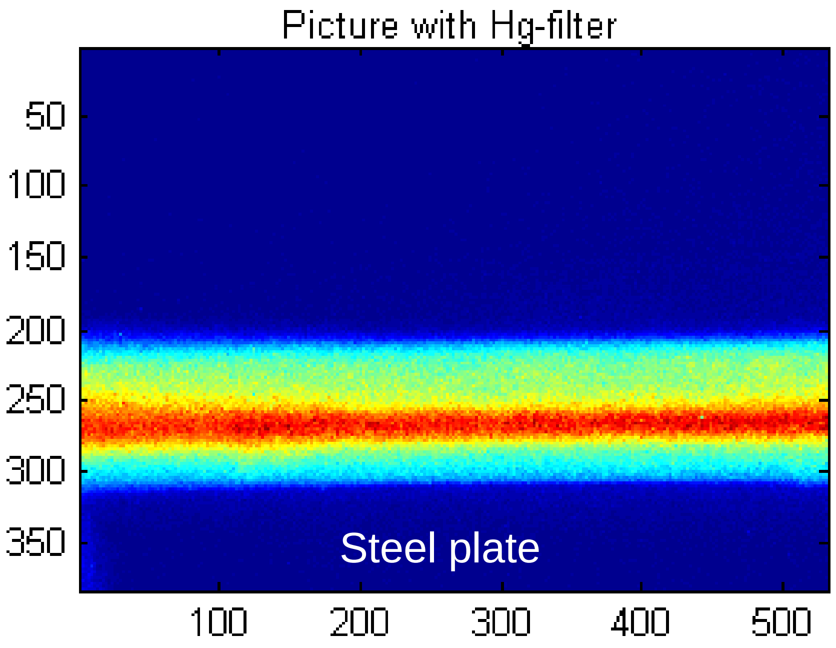


Specific experimental components are shown in figure:

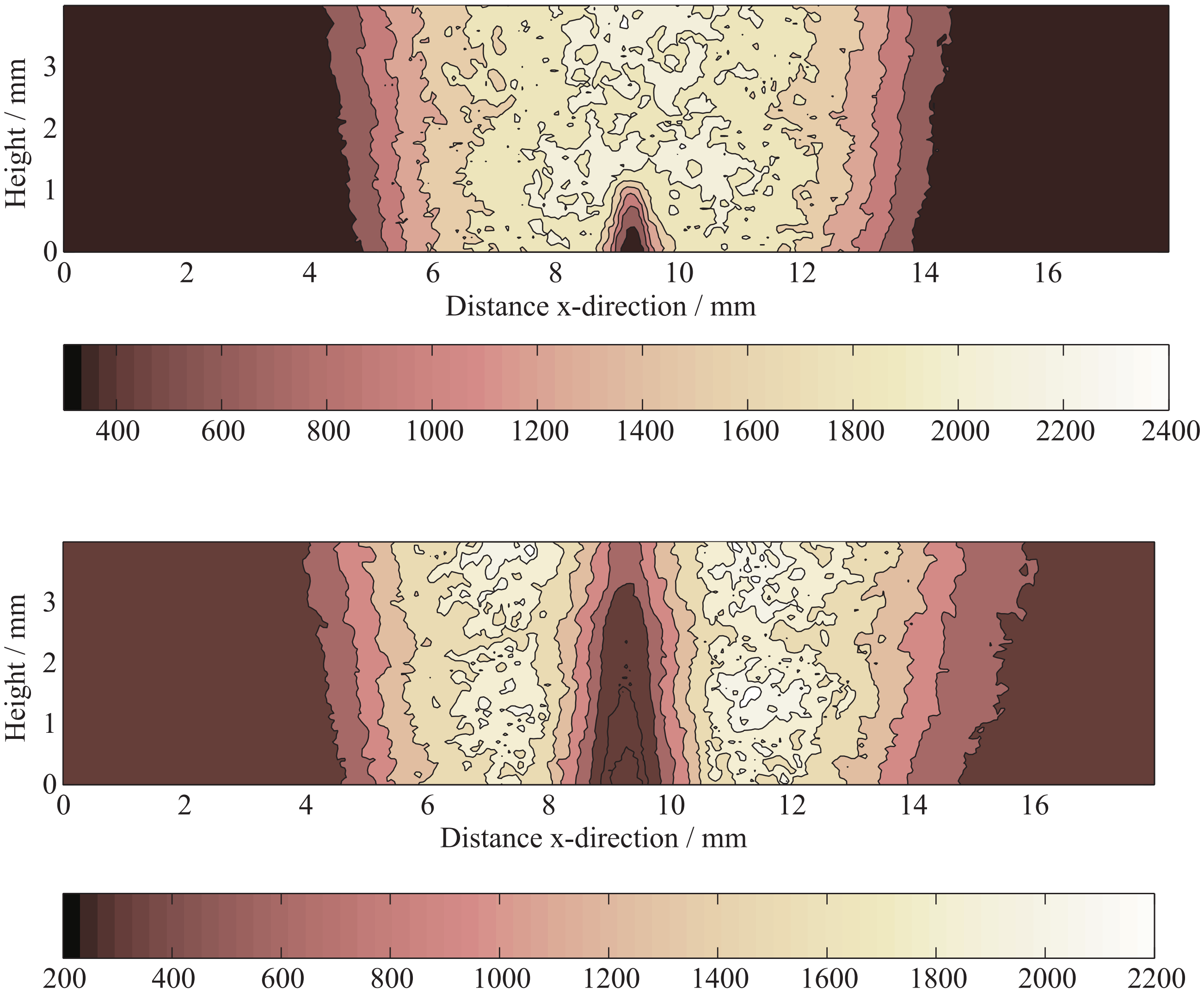
- Single-mode tunable laser
- Atomic (or molecular) gas filters

Influence of a Hg filter on the Rayleigh signal


Picture without Hg-filter


Picture with Hg-filter

FRS 2D temperature distribution in acetylene/air premixed flames. The upper picture at $\phi = 1.6$ and the lower at $\phi = 2.4$.

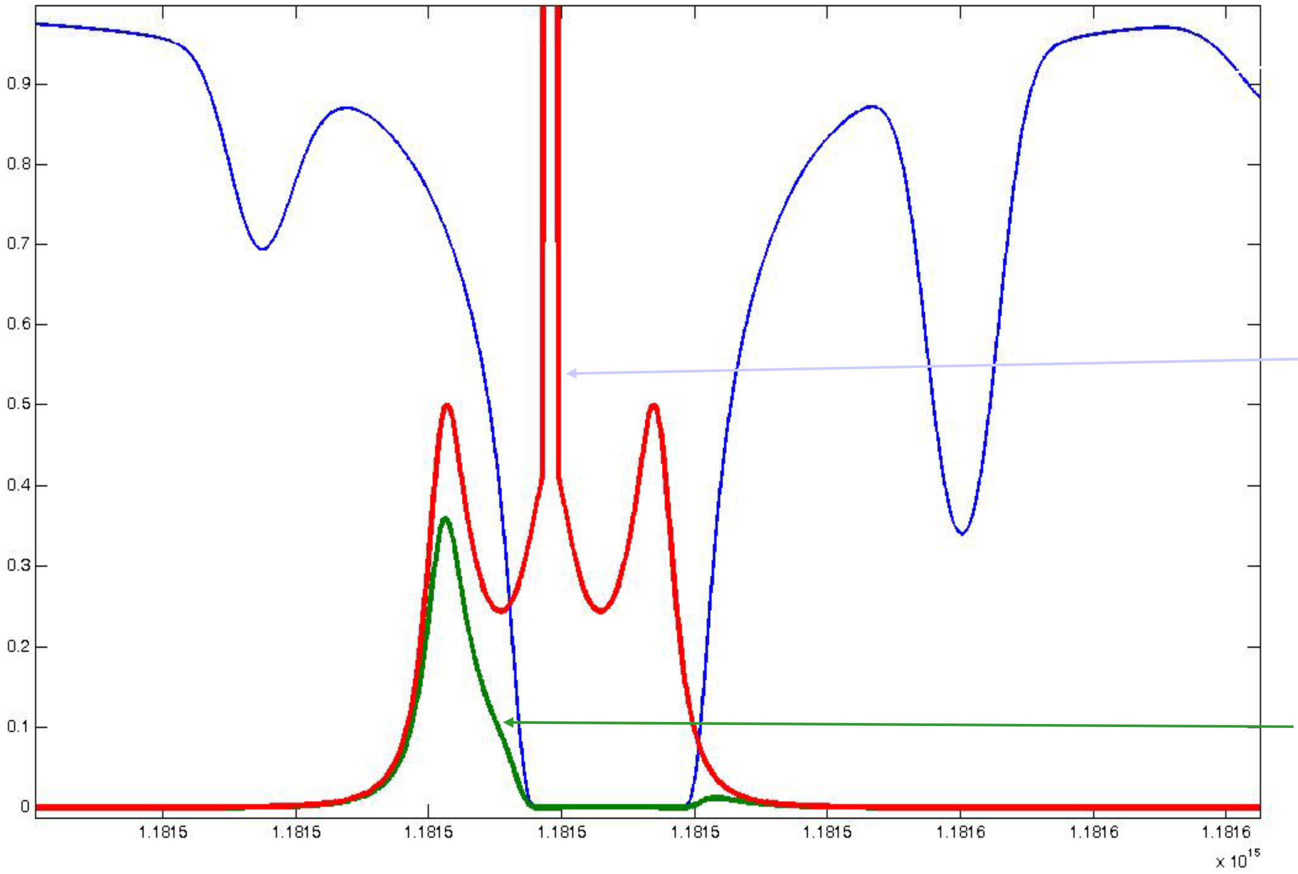


Filtered Rayleigh scattering: Fuel/air ratio imaging

- Rayleigh scattering cross-section: fuel molecules are much larger than air molecules
  - $\sigma_{propane} / \sigma_{air} \approx 15$
  - $\sigma_{isosctane} / \sigma_{air} \approx 90$
  - $\sigma_{diesel}/\sigma_{air} \approx 305$

- Spectrally resolved molecular Rayleigh scattering from spurious scattering light

Filtered Rayleigh scattering: lineshape (isooctane@20 bar, 800K)

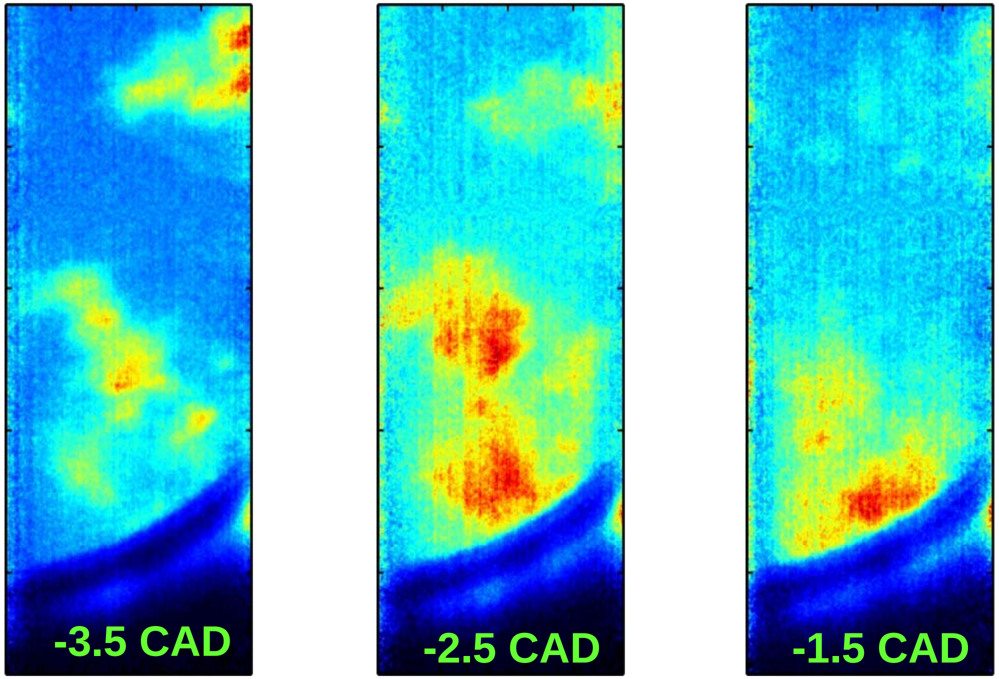


Hg filter transmission

Rayleigh scattering from isooctane and spurious scattering laser light

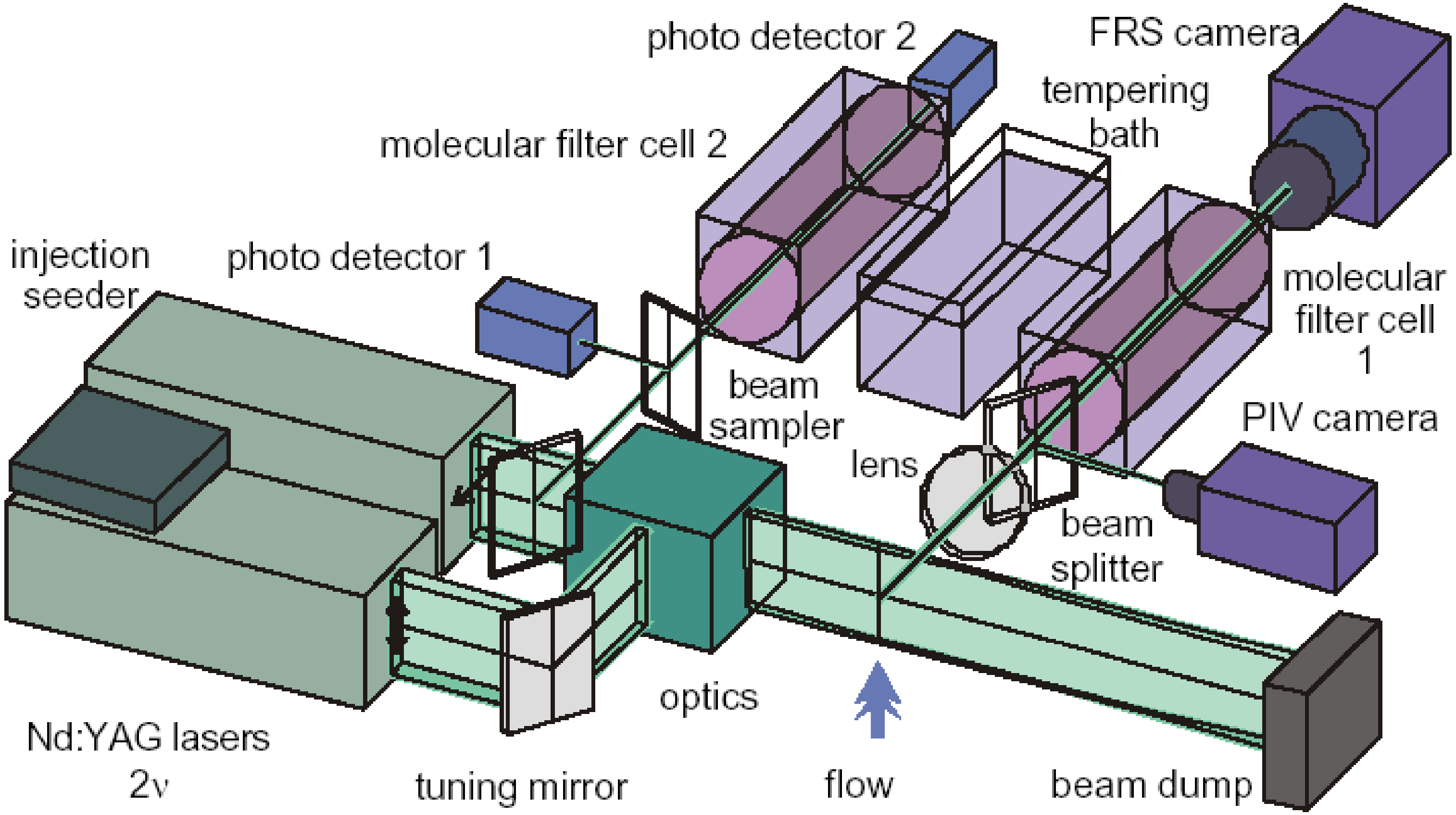
Transmitted FRS

Filtered Rayleigh scattering: Fuel/air ratio imaging in a diesel engine



Fuel injected at -10 CAD

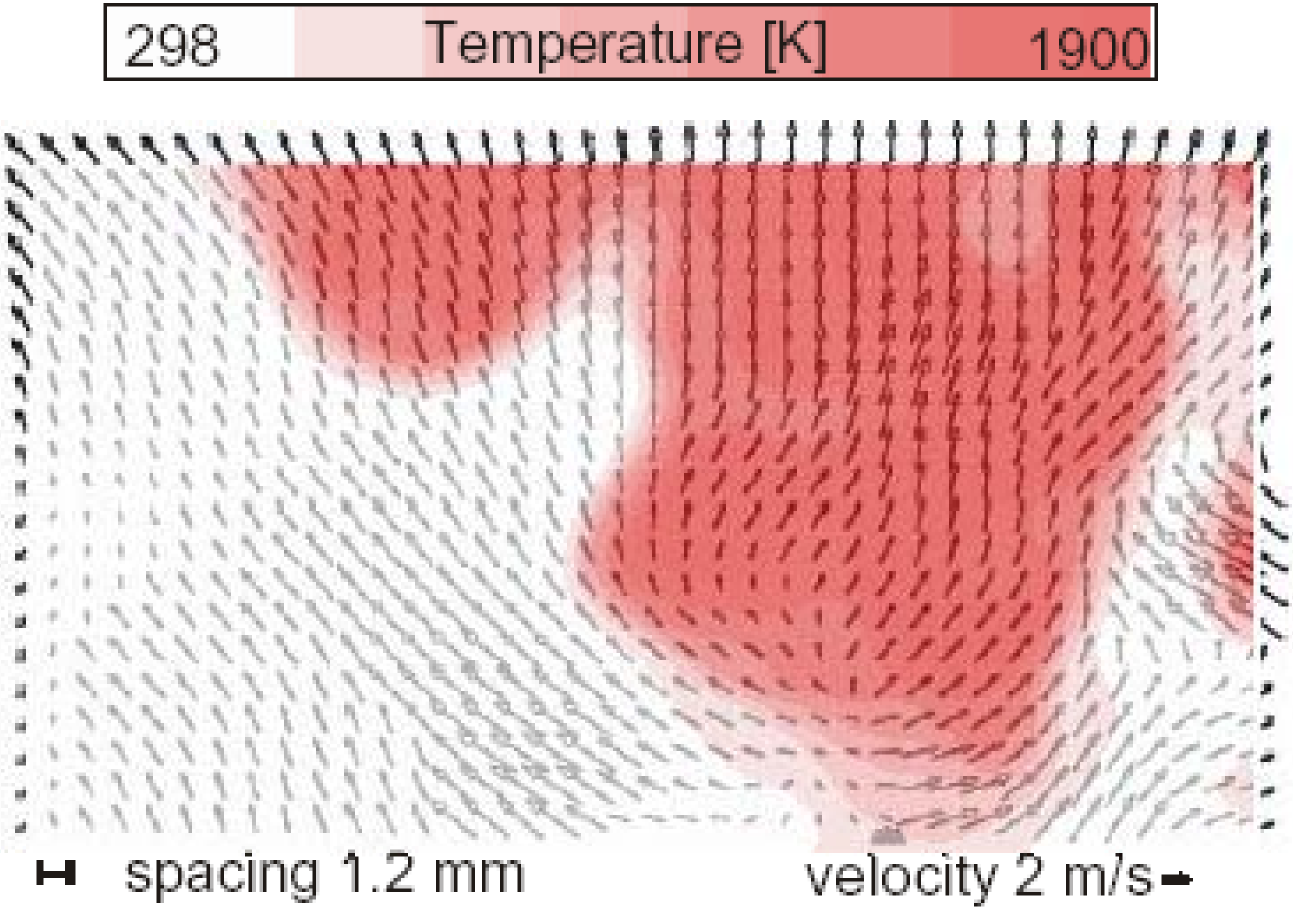
Simultaneous 2D FRS and PIV: Setup



Simultaneous thermometry using FRS and velocimetry using PIV.
The iodine filter absorb at the wavelength 532 nm.

D. Most and A. Leipertz, App. Opt. 40, 5379 (2001).

Simultaneous 2D FRS and PIV: Results



Instantaneous velocity and temperature field in a flame

D. Most and A. Leipertz, App. Opt. 40, 5379 (2001)

Summary of FRS

- FRS can be used in ’dirty’ environment and close to surfaces, since the scattering from surfaces, optics, droplets and particles give limited/no problem to the measurement.

- Major species concentrations have to be known or estimated for quantitative temperature measurements, because of the different Rayleigh scattering cross-sections for different species.

- The use of FRS in IC-engines has so far only been some initial demonstration measurements.

- FRS requires proper modeling of the lineshape, especially at high pressure and complex molecules (hydrocarbons)