

Lecture 31. Constituent Lidar (3)

Raman Lidar and Raman DIAL

- ❑ Introduction
- ❑ Fundamentals of Raman Scattering
- ❑ Raman Lidar Technical Requirements
- ❑ Raman Lidar Measurements of Water Vapor
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- ❑ Summary

Introduction

- ❑ Raman scattering is a very powerful spectroscopy tool to study many molecular structures and spectroscopy. As a result, the Raman lidar is very useful in constituent detection in the lower atmosphere. The best chapter describing Raman lidar is given by Dr. Ulla Wandinger of Leibniz Institute for Tropospheric Research, Germany. A scanned PDF of this chapter can be found at our class website.
- ❑ Raman lidars have come widely used instruments in atmospheric research, which have very low demands concerning spectral purity of the emitted laser light and frequency stabilization of the receiver. However, it suffers from the low cross sections of Raman scattering and thus from the comparably small signal-to-noise ratios of the measurements.
- ❑ Raman measurements do not require specific laser wavelengths. Because of the wavelength dependence of the Raman scattering cross-section which is proportional to λ^{-4} , shorter laser wavelengths are preferred. Attenuation of the laser light by gas absorption, especially ozone, can be avoided if wavelengths longer than 320 nm are chosen. However, the solar-blind region below 300 nm has also been used for Raman lidar. Concerning measurement range, wavelengths of 320–550 nm are best suited for Raman applications.

Introduction

Table 9.1. Overview of Raman lidar techniques. VRR – vibration–rotation Raman band, RR – rotational Raman band, SB – solar-blind spectral region

| Measured quantity | Interacting molecule, Raman band used | Typical achievable measurement range |
|--|---|---|
| Water-vapor mixing ratio | H ₂ O (vapor), VRR (+ reference gas, VRR) | 0–12 km (night) [16, 47] 0–5 km (day) [11, 47] |
| Extinction coefficient Backscatter coefficient Lidar ratio | N ₂ or O ₂ , VRR or RR (+ elastic signal for back- scatter and lidar ratio) | 0–30 km (night) [48] 0–10 km (day) [49] |
| Temperature | N ₂ and/or O ₂ , RR | 0–40 km (night) [19, 40] 0–12 km (day) [19] |
| Ozone concentration (Raman DIAL) | N ₂ and/or O ₂ , VRR-VRR or RR-VRR | 3–20 km (night) [46] 0–3 km (day SB) [29] |
| Other trace-gas concentrations | Specific gas, VRR (+ reference gas, VRR) | 0–1 km (day and night) [32] |
| Liquid water | H ₂ O (liquid), VRR (+ reference gas, VRR) | 0–4 km (night) [34] |

[Ulla Wandinger, Raman Lidar chapter, 2005]

Fundamentals of Raman Scattering

□ Raman scattering is an inelastic scattering and the Raman shift is characteristic for the scattering molecule. Using wavenumber $\tilde{\nu}=1/\lambda$, the Raman shift is given by

$$\Delta\tilde{\nu} = \tilde{\nu}_1 - \tilde{\nu}_s = \frac{\Delta E}{hc_0}$$

Where ΔE is the energy difference between the molecular energy levels involved in the Raman scattering.

Table 9.2. Molecular constants for the calculation of Raman frequency shifts of nitrogen and oxygen. The values for $\tilde{\nu}_{\text{vib}}$ consider an anharmonicity correction for the transition from the ground state to the first excited vibrational state (see Ref. [53], p. 125 and 182)

| Gas | $\tilde{\nu}_{\text{vib}}$ cm^{-1} | B_0 cm^{-1} | B_1 cm^{-1} | D_0 cm^{-1} |
|----------------|--|---------------------------|---------------------------|----------------------------|
| N ₂ | 2330.7 | 1.98957 [59] | 1.97219 [59] | 5.76×10^{-6} [59] |
| O ₂ | 1556.4 | 1.43768 [58] | 1.42188 [61] | 4.85×10^{-6} [58] |

Fundamentals of Raman Scattering

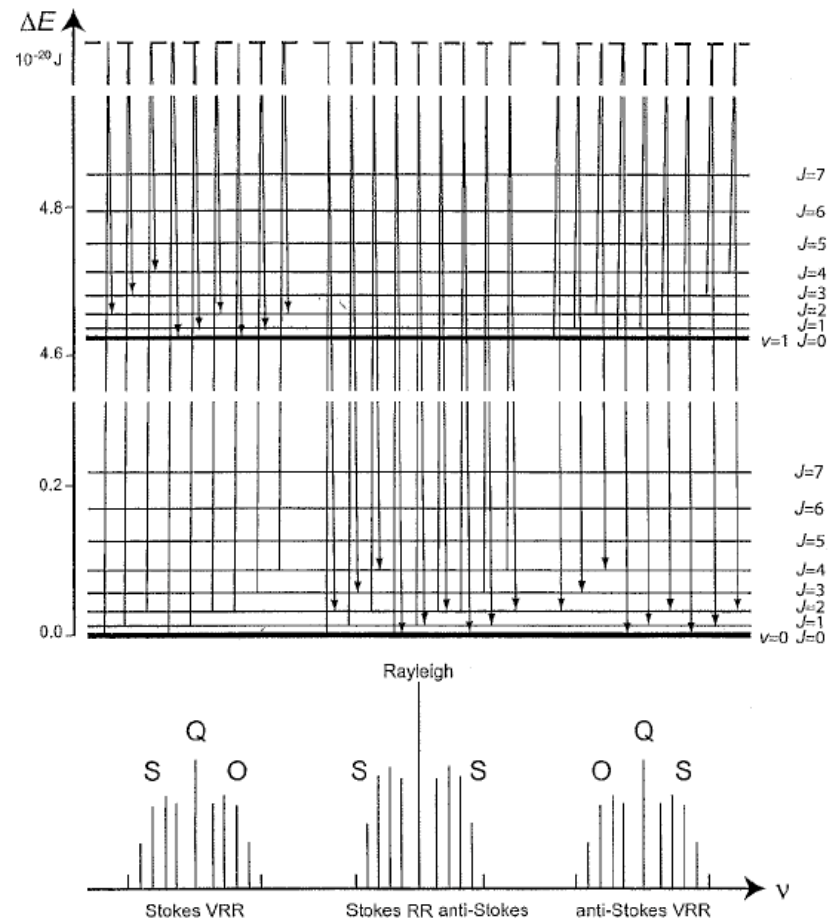


Fig. 9.1. Vibration-rotation energy levels of the N₂ molecule, Raman transitions, and resulting spectrum.

Selection Rules: $\Delta v = 0, \pm 1$ and $\Delta J = 0, \pm 2,$

Raman Lidar Technical Requirements

- ❑ Raman lidar has low Raman scattering cross section, so a high-power laser transmitter and a highly efficient receiving and detection system are required.
- ❑ Laser: no specific wavelength is needed, but ideal wavelengths are between 320 and 550 nm
- ❑ Beam expander: is to reduce the laser beam divergence in order to allow for a narrow telescope field of view, which will help to suppress background light and increase SNR. Usually go from 1 mrad to 0.1 mrad.
- ❑ Telescope: usually 0.5–1 m in diameter
- ❑ Receiver optics:
- ❑ Detectors and DAQ:

Water Vapor Raman Lidar Setup

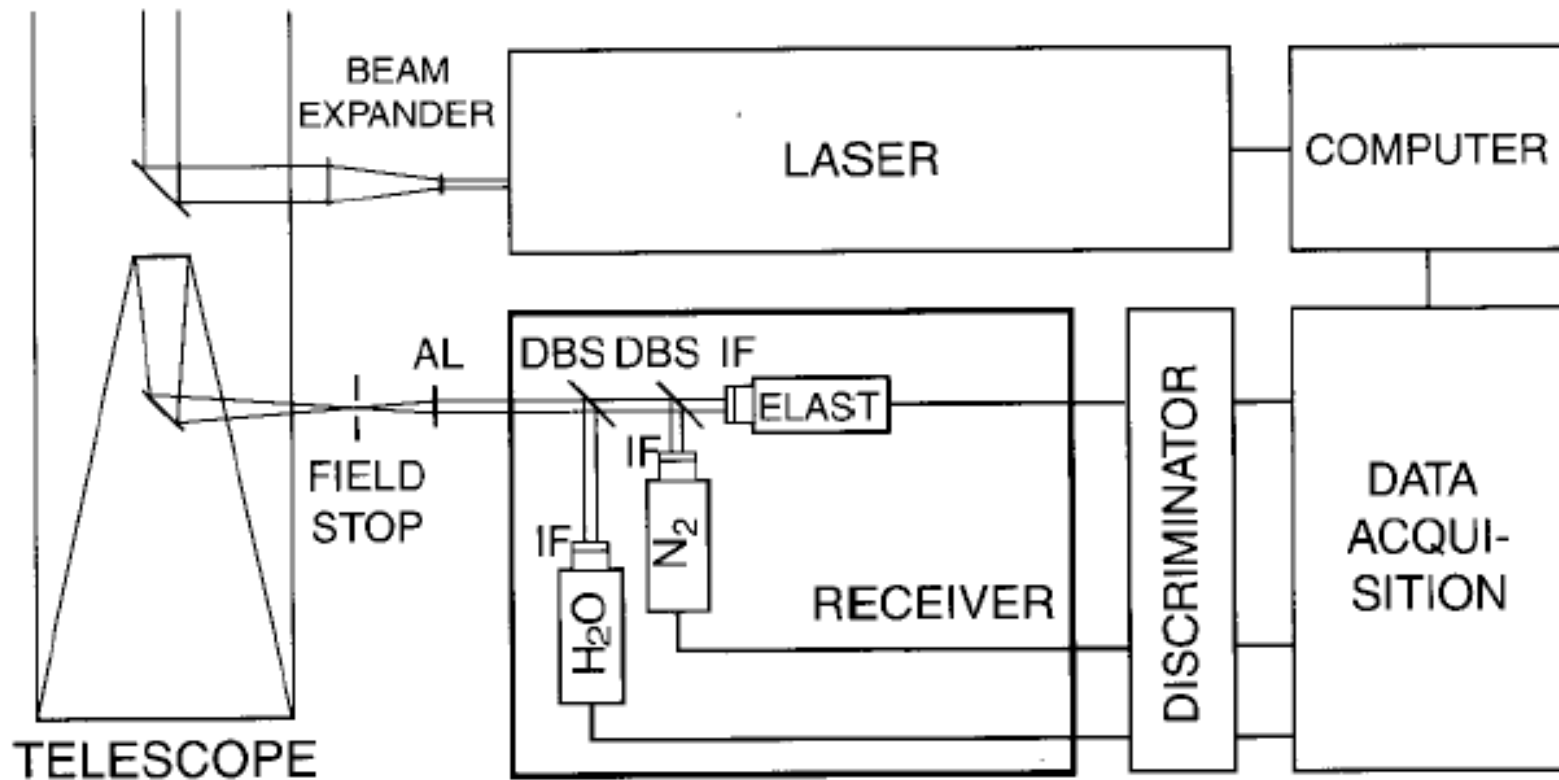
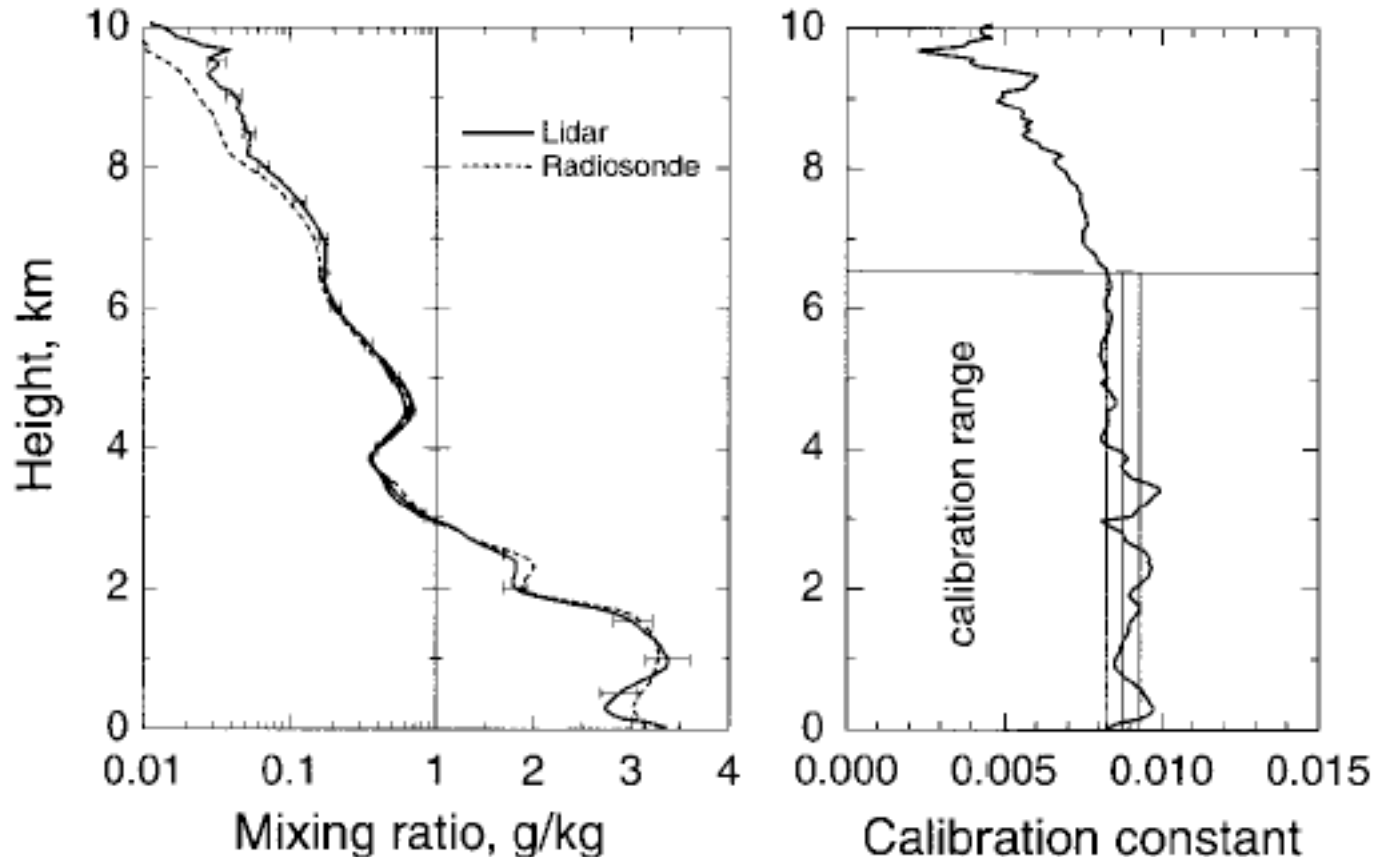


Fig. 9.3. Typical setup of a water-vapor and aerosol Raman lidar. AL – achromatic lens, IF – interference filter, DBS – dichroic beam splitter.

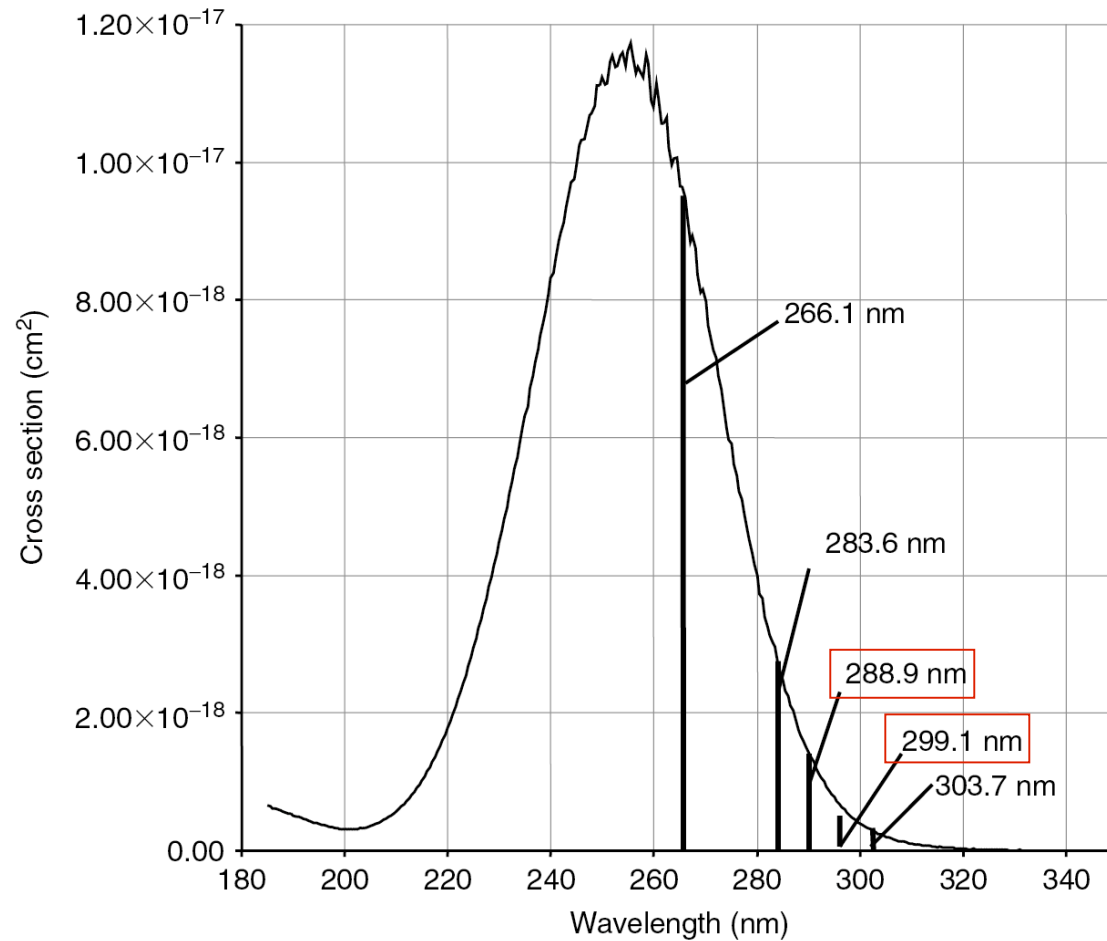
- ❑ Mixing ratio: two Raman channels: one is the return from the gas of interest and another is the return of a reference gas (N₂ or O₂).

Water Vapor Results by Raman Lidar



- ❑ Raman scattering lidar usually needs calibration with radiosonde or independent calibration by measuring or calculating the relevant system parameters or by using multichannel to measure the aerosol extinction and effective cross-sections.

Conventional DIAL Ozone Measurement



$$\lambda_{ON} = 288.9nm$$

$$\lambda_{OFF} = 299.1nm$$

Figure 4.1 Ozone absorption spectrum in the UV: the Raman shifted wavelengths given in Table 4.2 are highlighted in this figure.

Conventional DIAL Solutions

$$n_c(R) = \frac{1}{2\Delta\sigma_{abs}} \frac{d}{dR} \left\{ \begin{aligned} & \ln \left[\frac{P_S(\lambda_{OFF}, R) - P_B}{P_S(\lambda_{ON}, R) - P_B} \right] \\ & - \ln \left[\frac{P_L(\lambda_{OFF})\eta(\lambda_{OFF})}{P_L(\lambda_{ON})\eta(\lambda_{ON})} \right] \\ & - \ln \left[\frac{\beta_{aer}(\lambda_{OFF}, R) + \beta_{mol}(\lambda_{OFF}, R)}{\beta_{aer}(\lambda_{ON}, R) + \beta_{mol}(\lambda_{ON}, R)} \right] \end{aligned} \right\} \\
- \frac{1}{\Delta\sigma_{abs}} \left\{ \begin{aligned} & [\alpha_{aer}(\lambda_{ON}, R) - \alpha_{aer}(\lambda_{OFF}, R)] \\ & + [\alpha_{mol}(\lambda_{ON}, R) - \alpha_{mol}(\lambda_{OFF}, R)] \\ & + [\sigma_{IG}(\lambda_{ON}, R) - \sigma_{IG}(\lambda_{OFF}, R)]n_{IG} \end{aligned} \right\}$$

All terms non-range dependent vanish after the derivative d/dR

Introducing large error when large aerosol gradient or IG exists

Ozone Raman DIAL

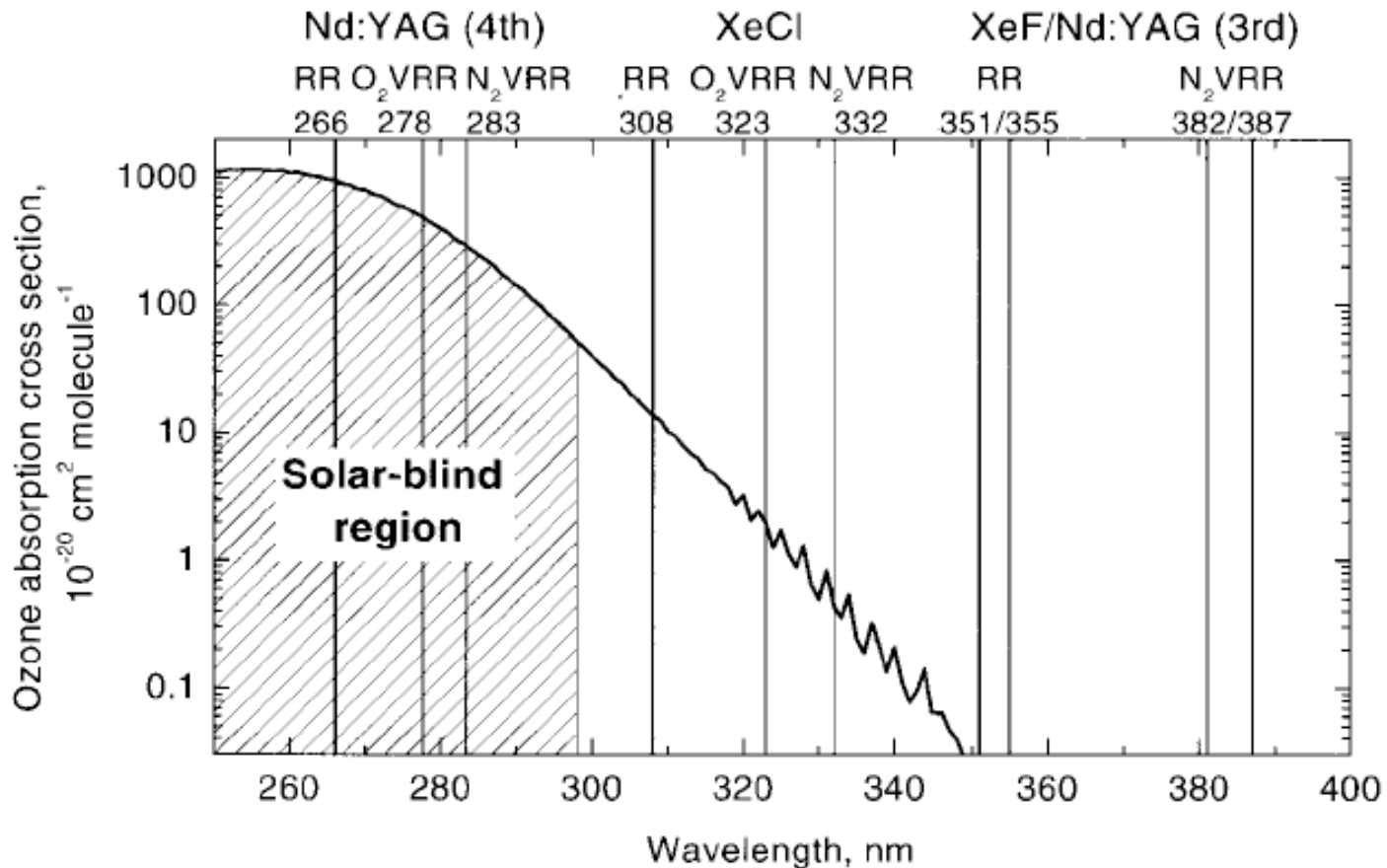


Fig. 9.5. Ozone absorption cross section and wavelengths applied in the ozone Raman DIAL technique for upper tropospheric and stratospheric measurements ($\lambda > 300 \text{ nm}$) and for boundary-layer measurements in the solar-blind region ($\lambda < 300 \text{ nm}$).

[Ulla Wandinger, Raman Lidar chapter, 2005]

Raman DIAL Equations

□ Raman channels (e.g., from N₂ Raman inelastic scattering, instead of elastic scattering from air molecules or aerosols),

$$\begin{aligned}
 P_S(\lambda_{\text{Ref}}^{\text{ON}}, R) &= P_L(\lambda_0^{\text{ON}}) \left[\beta_{\text{Raman}}(\lambda_0^{\text{ON}}, \lambda_{\text{Ref}}^{\text{ON}}, R) \Delta R \right] \left(\frac{A}{R^2} \right) \\
 &\times \exp \left[- \int_0^R \left(\alpha_{\text{aer}}(\lambda_0^{\text{ON}}, r) + \alpha_{\text{aer}}(\lambda_{\text{Ref}}^{\text{ON}}, r) + \alpha_{\text{mol}}(\lambda_0^{\text{ON}}, r) + \alpha_{\text{mol}}(\lambda_{\text{Ref}}^{\text{ON}}, r) \right) dr \right] \\
 &\times \exp \left[- \int_0^R \left(\sigma_{\text{IG}}(\lambda_0^{\text{ON}}, r) + \sigma_{\text{IG}}(\lambda_{\text{Ref}}^{\text{ON}}, r) \right) n_{\text{IG}} dr \right] \\
 &\times \exp \left[- \int_0^R \left(\sigma_{\text{abs}}(\lambda_0^{\text{ON}}, r) + \sigma_{\text{abs}}(\lambda_{\text{Ref}}^{\text{ON}}, r) \right) n_c(r) dr \right] \left[\eta(\lambda_{\text{Ref}}^{\text{ON}}) G(R) \right] + P_B
 \end{aligned}$$

$$\begin{aligned}
 P_S(\lambda_{\text{Ref}}^{\text{OFF}}, R) &= P_L(\lambda_0^{\text{OFF}}) \left[\beta_{\text{Raman}}(\lambda_0^{\text{OFF}}, \lambda_{\text{Ref}}^{\text{OFF}}, R) \Delta R \right] \left(\frac{A}{R^2} \right) \\
 &\times \exp \left[- \int_0^R \left(\alpha_{\text{aer}}(\lambda_0^{\text{OFF}}, r) + \alpha_{\text{aer}}(\lambda_{\text{Ref}}^{\text{OFF}}, r) + \alpha_{\text{mol}}(\lambda_0^{\text{OFF}}, r) + \alpha_{\text{mol}}(\lambda_{\text{Ref}}^{\text{OFF}}, r) \right) dr \right] \\
 &\times \exp \left[- \int_0^R \left(\sigma_{\text{IG}}(\lambda_0^{\text{OFF}}, r) + \sigma_{\text{IG}}(\lambda_{\text{Ref}}^{\text{OFF}}, r) \right) n_{\text{IG}} dr \right] \\
 &\times \exp \left[- \int_0^R \left(\sigma_{\text{abs}}(\lambda_0^{\text{OFF}}, r) + \sigma_{\text{abs}}(\lambda_{\text{Ref}}^{\text{OFF}}, r) \right) n_c(r) dr \right] \left[\eta(\lambda_{\text{Ref}}^{\text{OFF}}) G(R) \right] + P_B
 \end{aligned}$$

Solution for Raman DIAL Equations

From the two Raman reference channel equations, we obtain the number density of the constituent that we are interested in

$$n_c(R) = \frac{1}{\Delta\sigma_{abs}} \frac{d}{dR} \left\{ \begin{array}{l} \ln \left[\frac{P_S(\lambda_{Ref}^{OFF}, R) - P_B}{P_S(\lambda_{Ref}^{ON}, R) - P_B} \right] \\ - \ln \left[\frac{P_L(\lambda_0^{OFF}) \eta(\lambda_{Ref}^{OFF})}{P_L(\lambda_0^{ON}) \eta(\lambda_{Ref}^{ON})} \right] \\ - \ln \left[\frac{\beta_{Raman}(\lambda_{Ref}^{OFF}, R)}{\beta_{Raman}(\lambda_{Ref}^{ON}, R)} \right] \\ \Delta\alpha_{aer}(R) \\ + \Delta\alpha_{mol}(R) \\ + \Delta\sigma_{IG}(R)n_{IG} \end{array} \right\}$$

A
B
C
D
E
F

Here, the Δ expressions consist of four terms each

$$\Delta\xi = \xi(\lambda_0^{ON}) + \xi(\lambda_{Ref}^{ON}) - \xi(\lambda_0^{OFF}) - \xi(\lambda_{Ref}^{OFF})$$

with

$$\xi = \sigma_{abs}, \alpha_{aer}, \alpha_{mol}, \sigma_{IG}$$

Solution in Ozone Case

- ❑ Term B can be measured and it is range-independent, so the derivative is zero,
- ❑ Term C is only concerned about molecule Raman scattering, so can be calculated.
- ❑ Term D will be determined through using the Raman channel at OFF wavelength and introducing Angstrom exponent.
- ❑ Term E is concerned about molecule Rayleigh scattering, so can be calculated from atmosphere temperature and pressure.
- ❑ Term F can be minimized through choosing proper wavelengths, thus, can be ignored.

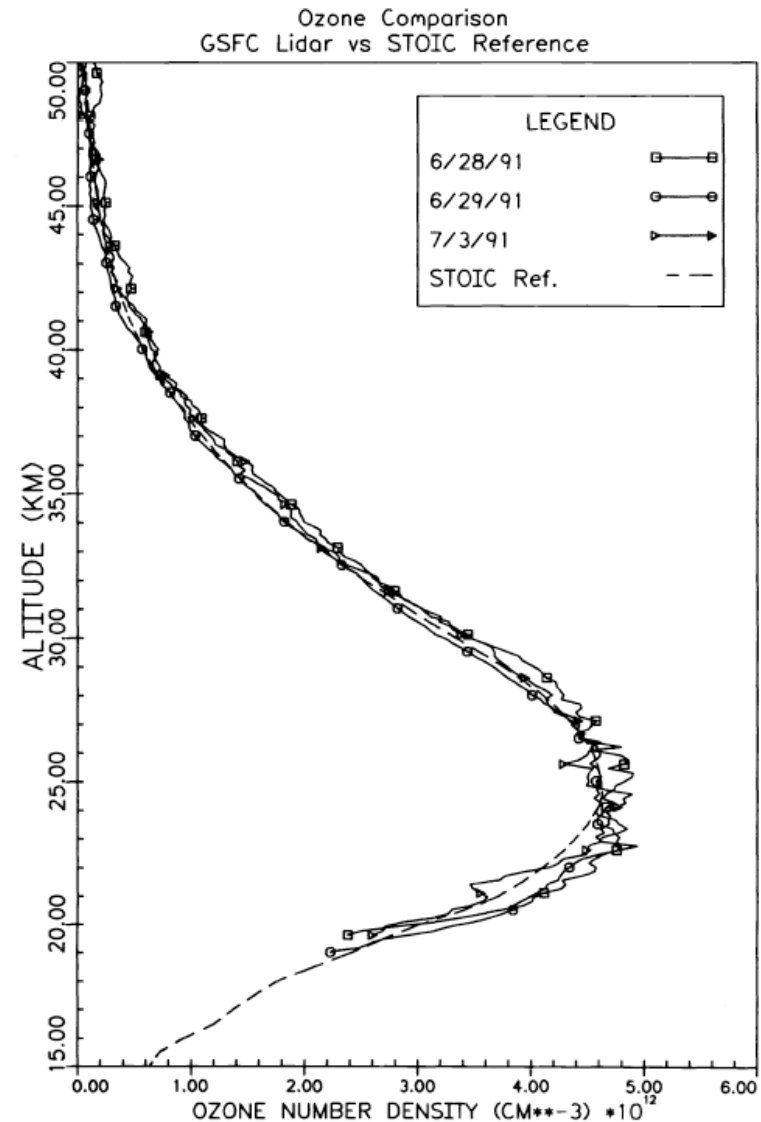
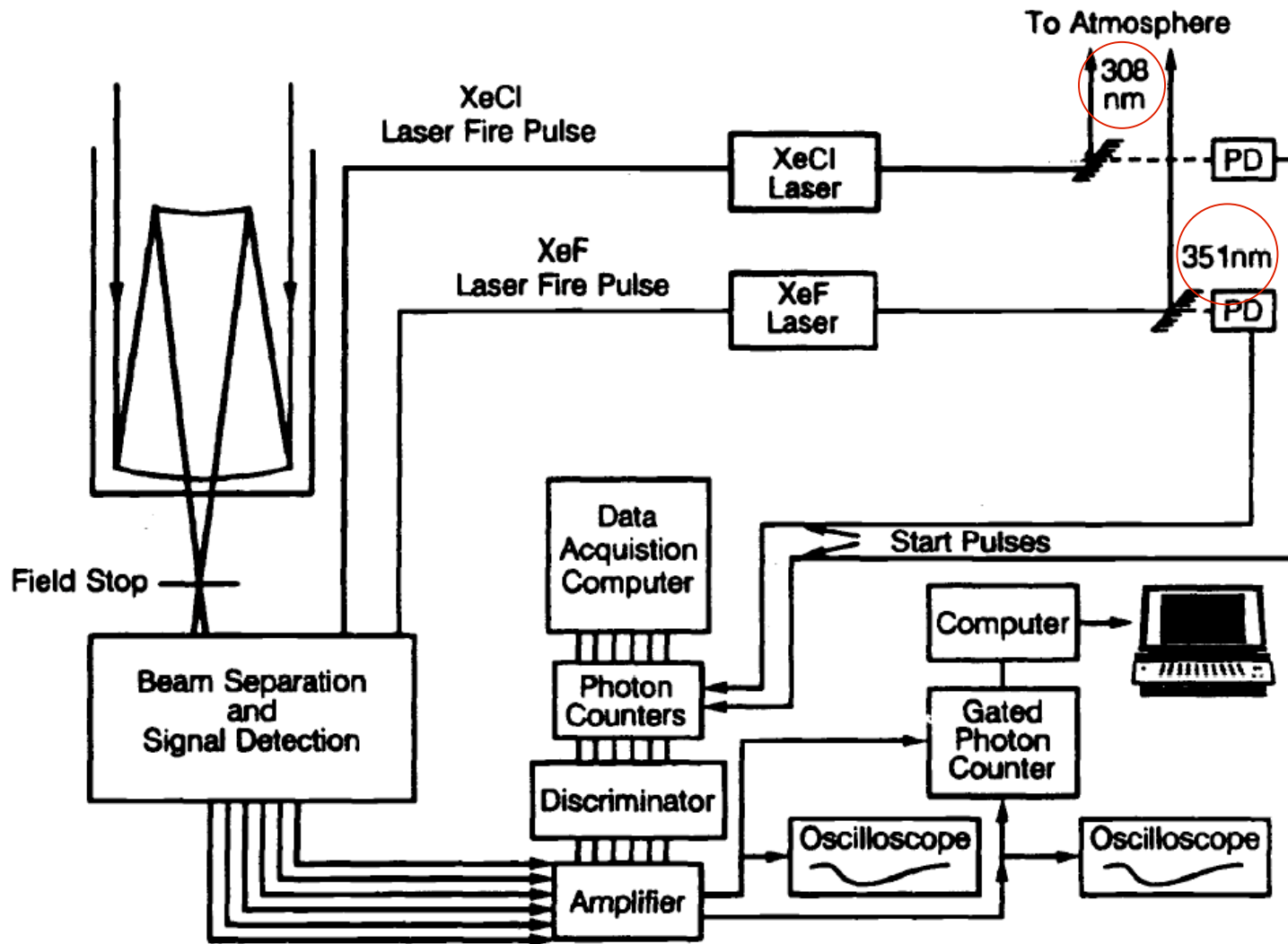


Fig. 6 GSFC lidar ozone profiles taken at JPL-TMO during June and July 1991 using the mechanical chopper.

Ozone Raman DIAL Instrumentation

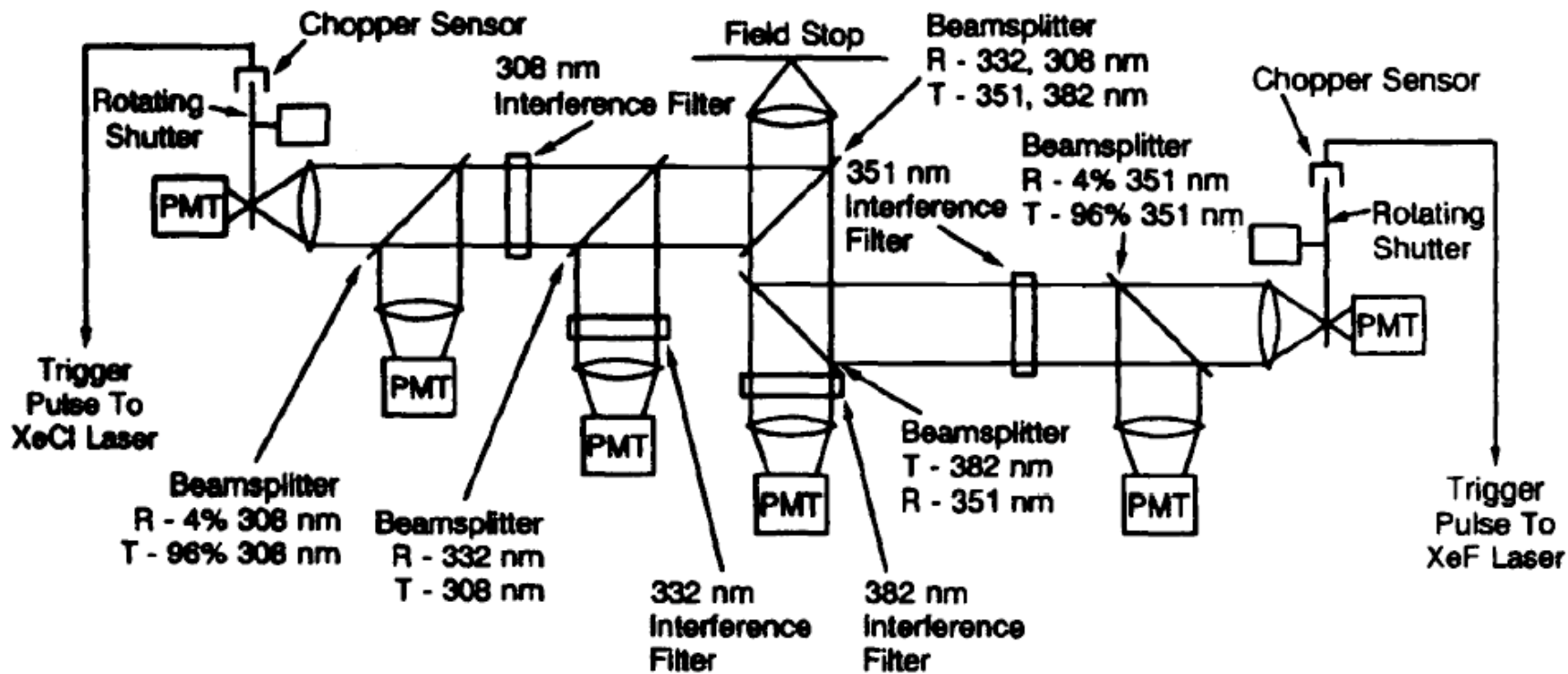


Return Signals -

On-line: 332 nm (N_2 Raman scattering from 308 nm)

Off-line: 382 nm (N_2 Raman scattering from 351 nm)

Ozone Raman DIAL Receiver



Summary

- ❑ The essential point for Raman lidar is to avoid the aerosol scattering in the Raman-shifted channel. Thus, only aerosol extinction will be dealt with in deriving constituent density. Aerosol extinction can be safely estimated by introducing Angstrom exponent. The error introduced by Angstrom uncertainty is much less than the lidar ratio or backscatter coefficient.
- ❑ Combination of Raman and DIAL can effectively remove the influence from aerosol or interference gases.
- ❑ More combination and approaches using DIAL and Raman are on the way to solve practical constituent detection problems.
- ❑ Again, spectroscopy is the key to guide the laser wavelength selection and DIAL/Raman considerations.