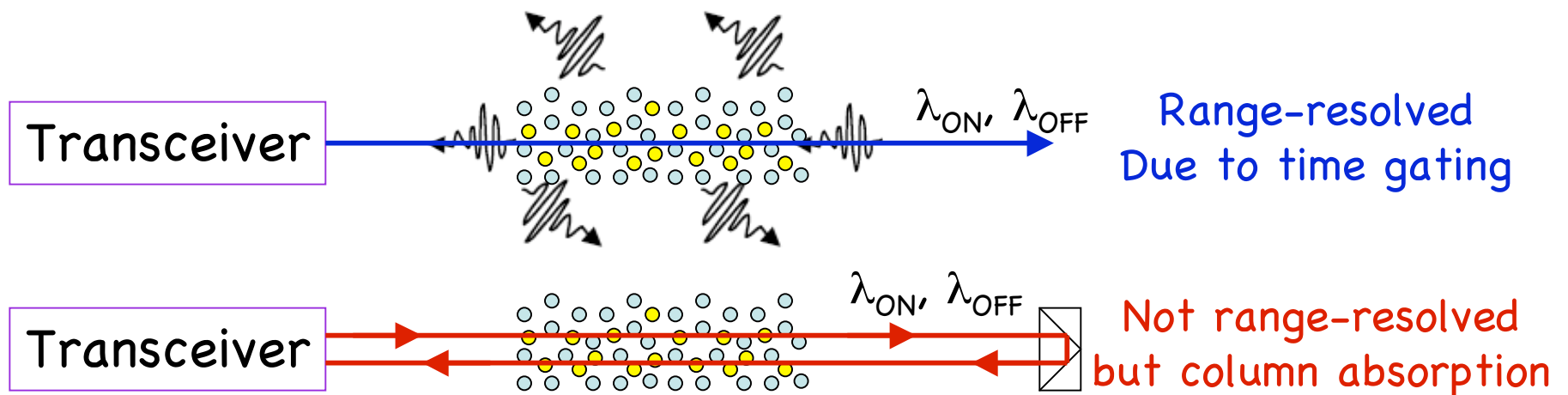


Lecture 28. Constituent Lidar (2)

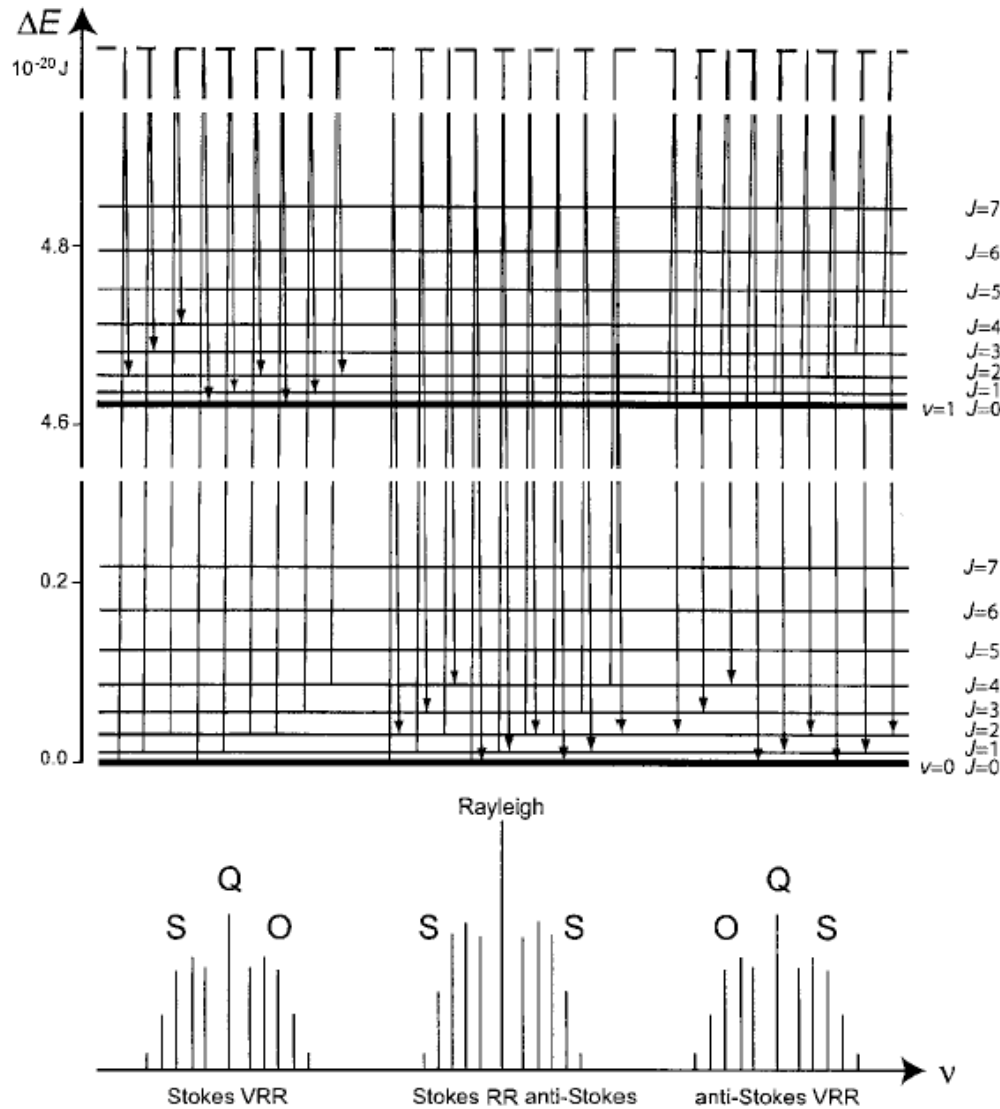
- Review of Constituent Lidars
- Raman Scattering Lidar for Constituent Detection
(Example: Water Vapor Measurement)
- Raman DIAL
(Example: Ozone Measurement)
- RVR Raman DIAL
- Summary

Review of Constituent Measurements

- ❑ Importance of constituent study
- ❑ Lidar detection of constituents - key points
- ❑ Resonance fluorescence lidar technique
- ❑ DIAL technique



Raman Scattering (Inelastic)



$$\Delta k = k_i - k_s = \frac{\Delta E}{hc}$$

$$k = \frac{1}{\lambda}$$

Wavenumber

Fig. 9.1. Vibration–rotation energy levels of the N_2 molecule, Raman transitions, and resulting spectrum.

Raman Scattering Frequency Shifts

- ❑ The scattering of incident radiation by atmospheric molecules involves both elastic and inelastic processes. (Wavenumber $k = 1/\lambda$)
- ❑ Elastic or Rayleigh (Cabannes) scattering is the scattering that the frequency of the scattered photon k_s is the same as the frequency of the incident photon k_i . In this case the molecule preserves its vibration-rotation energy level during the scattering process.
- ❑ Inelastic or Raman scattering processes lead to a change of the molecule's quantum state, and a change of the frequency of the scattered photon with respect to the incident photon frequency.
- ❑ If the molecule absorbs energy and transits to a higher energy level, the frequency of the scattered photon is decreased, so the wavelength is red-shifted. This is Stokes Raman scattering.
- ❑ If the molecule transfers energy to the scattered photon by decreasing its energy level, the frequency of the scattered photon is increased, so the wavelength is blue-shifted. This is anti-Stokes Raman scattering.
- ❑ The Raman frequency shift is $\Delta k = k_i - k_s = \Delta E/(hc)$
- ❑ Raman scattering cross-section is proportional to λ^{-4} .

Raman Backscatter Spectrum

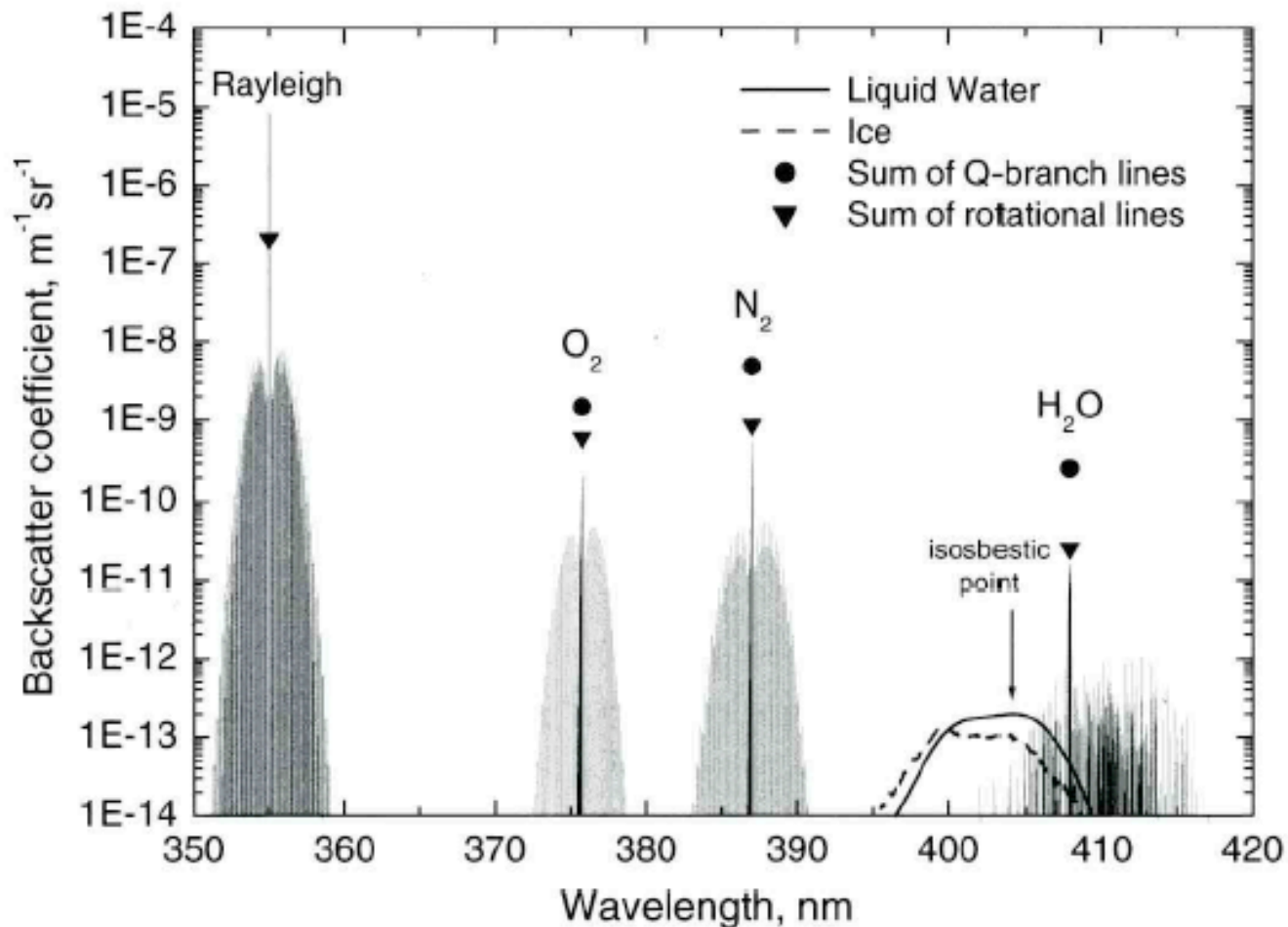


Fig. 9.2. Raman backscatter spectrum of the atmosphere for an incident laser wavelength of 355 nm, normal pressure, a temperature of 300 K, an N₂ and O₂ content of 0.781 and 0.209, respectively, and a water-vapor mixing ratio of 10 g/kg. The curves for liquid water and ice are arbitrarily scaled. The isosbestic point is discussed in Subsection 9.5.2.

Water Vapor Raman Lidar Setup

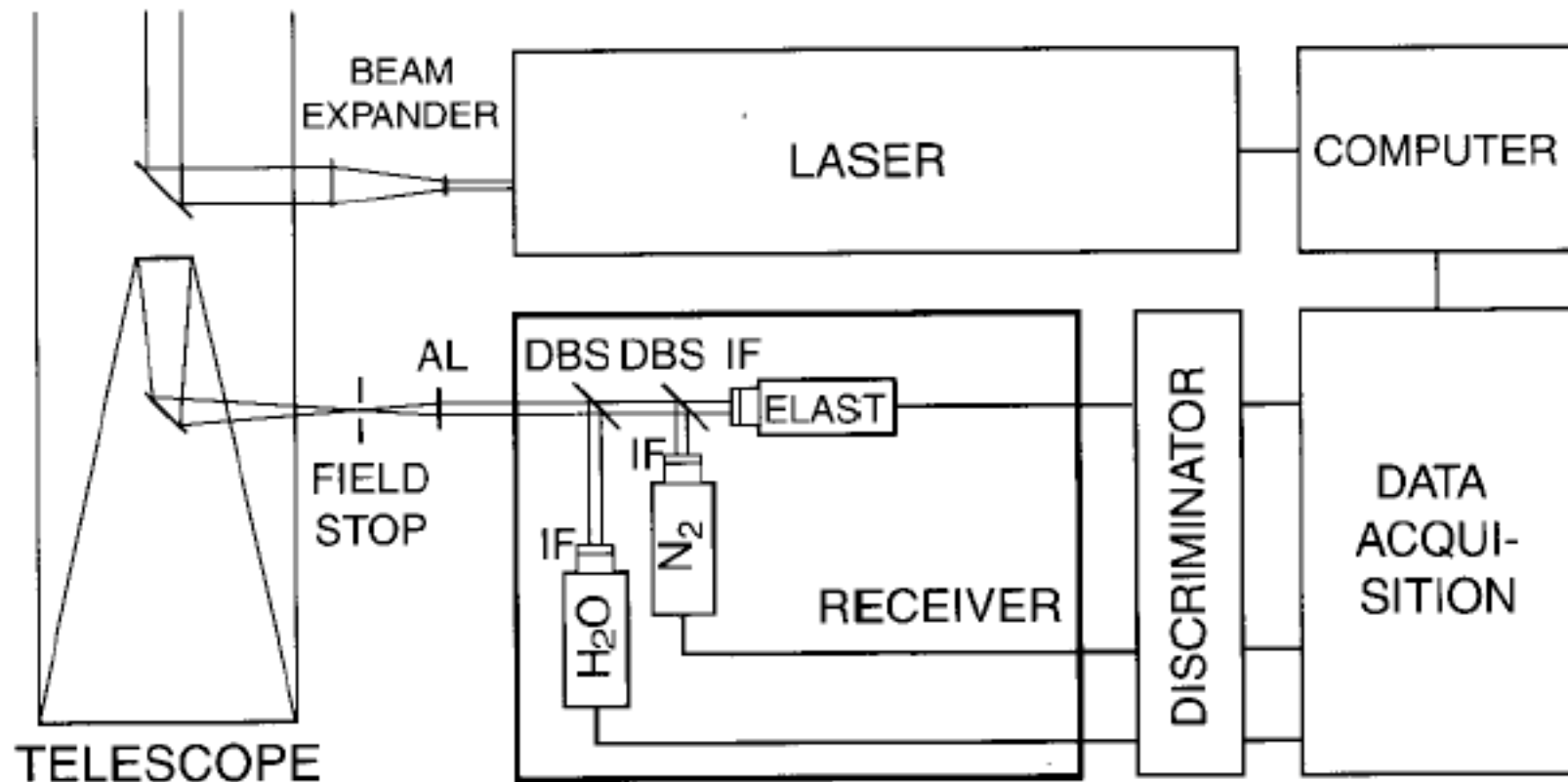


Fig. 9.3. Typical setup of a water-vapor and aerosol Raman lidar. AL – achromatic lens, IF – interference filter, DBS – dichroic beam splitter.

Raman Lidar Equation

- From Lecture 26, we rewrite the Raman lidar equation for the Raman scattering from the constituent interested and from the reference nitrogen N_2 or oxygen O_2

$$P_{cRa}(R, \lambda_{cRa}) = \frac{E_o \eta_{\lambda_{cRa}}}{R^2} O(R, \lambda_{cRa}) \beta_{cRa}(R, \lambda_0) \exp\left\{-\int_0^R [\alpha(r, \lambda_0) + \alpha(r, \lambda_{cRa})] dr\right\}$$

Extinction On the way up and on the way back

$$P_{RefRa}(R, \lambda_{RefRa}) = \frac{E_o \eta_{\lambda_{RefRa}}}{R^2} O(R, \lambda_{RefRa}) \beta_{RefRa}(R, \lambda_0) \exp\left\{-\int_0^R [\alpha(r, \lambda_0) + \alpha(r, \lambda_{RefRa})] dr\right\}$$

- The Raman backscatter coefficients from the constituent interested and from the reference atmosphere molecule can be written as

$$\beta_{cRa}(R, \lambda_0) = N_{cRa}(R) \frac{d\sigma_{cRa}}{d\Omega}(\pi, \lambda_0)$$

Constituent number density

$$\beta_{RefRa}(R, \lambda_0) = N_{RefRa}(R) \frac{d\sigma_{RefRa}}{d\Omega}(\pi, \lambda_0)$$

Reference molecule number density

Raman backscatter cross-section

Solution for Raman Lidar Equation

- Taking the ratio of the above two Raman lidar equations

$$\frac{P_{cRa}(R, \lambda_{cRa})}{P_{RefRa}(R, \lambda_{RefRa})} = \frac{\eta_{\lambda_{cRa}} N_{cRa}(R) \frac{d\sigma_{cRa}}{d\Omega}(\pi, \lambda_0) \exp\left\{-\int_0^R \alpha(r, \lambda_{cRa}) dr\right\}}{\eta_{\lambda_{RefRa}} N_{RefRa}(R) \frac{d\sigma_{RefRa}}{d\Omega}(\pi, \lambda_0) \exp\left\{-\int_0^R \alpha(r, \lambda_{RefRa}) dr\right\}}$$

Here, E_0 , R^2 , $O(R)$, and $\alpha(r, \lambda_0)$ are already cancelled out

- Thus, the mixing ratio of the constituent in the atmosphere is given by

$$r_m(R) = \frac{M_c(R)}{M_{air}(R)} = \frac{M_c(R)}{M_{Ref}(R)} \frac{M_{Ref}}{M_{air}} = \frac{M_{Ref}}{M_{air}} \frac{m_c}{m_{Ref}} \frac{N_{cRa}(R)}{N_{RefRa}(R)}$$

$$= \frac{M_{Ref}}{M_{air}} \frac{m_c}{m_{Ref}} \underbrace{\eta_{\lambda_{RefRa}} \frac{d\sigma_{RefRa}}{d\Omega}(\pi, \lambda_0)}_{\text{Calibration Constant } C} \frac{P_{cRa}(R, \lambda_{cRa}) \exp\left\{-\int_0^R \alpha(r, \lambda_{RefRa}) dr\right\}}{\eta_{\lambda_{cRa}} \frac{d\sigma_{cRa}}{d\Omega}(\pi, \lambda_0) P_{RefRa}(R, \lambda_{RefRa}) \exp\left\{-\int_0^R \alpha(r, \lambda_{cRa}) dr\right\}}$$

Calibration Constant C

Solution Cont'd

❑ The calibration of Raman lidar data lies in several aspects:

- (1) The ratio of reference molecule density in the air
- (2) The lidar detector efficiency ratio at two wavelengths
- (3) The effective Raman backscatter cross-section ratio
- (4) Atmospheric transmission ratio, i.e., extinction coefficient difference

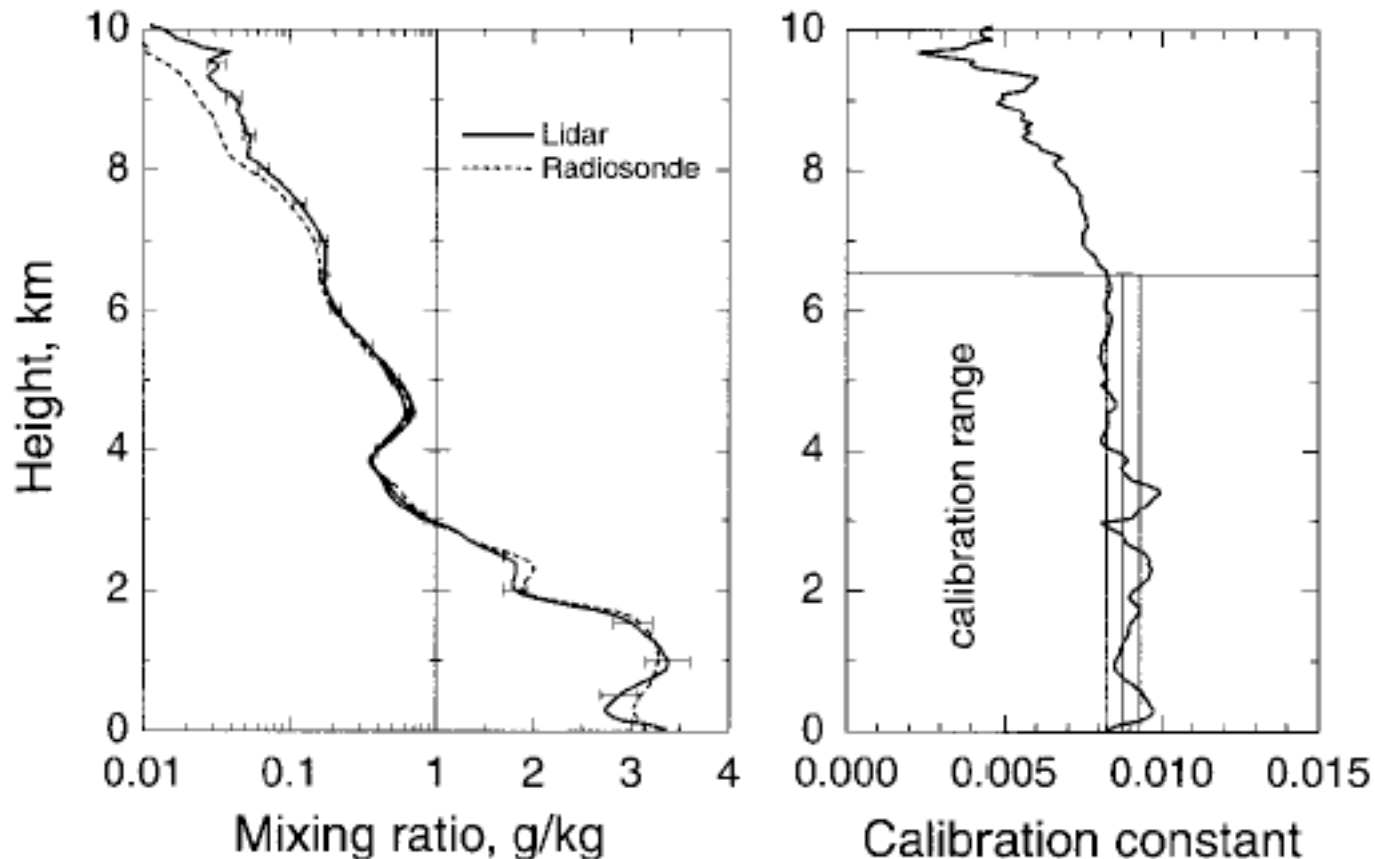
❑ Depending on the filter bandwidth, we have to take the average cross-section over the spectrum of the observed Raman band, weighted with the instrument's transmission function. It is also possible that we have to consider temperature dependence of the signal intensity distribution in the wings of the Raman bands.

❑ Regarding extinction coefficient at different wavelengths, it contains the extinction from aerosols and air molecules. Again, air molecules can be calculated from model data. For aerosols, we will introduce Angstrom exponent as we did in Lecture 26. This factor $a(R)$ is usually between 0-2, and normally 1. The error introduced by Angstrom factor is less than 10%.

$$\alpha(R) = \alpha_{aer}(R) + \alpha_{mol}(R)$$

$$\frac{\alpha_{aer}(\lambda_0)}{\alpha_{aer}(\lambda_{Ra})} = \left(\frac{\lambda_{Ra}}{\lambda_0} \right)^{a(R)}$$

Raman Water Vapor Results



- ❑ 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.

Ozone Measurements

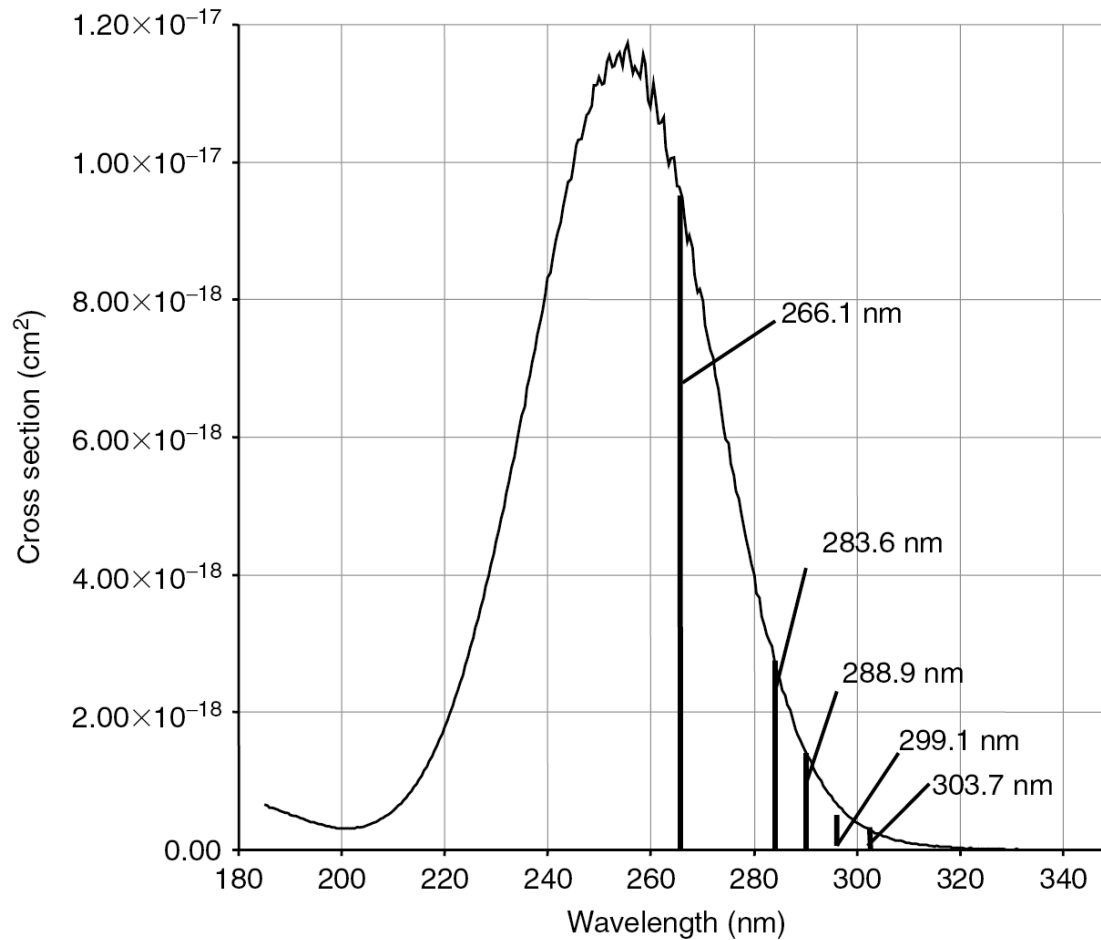


Figure 4.1 Ozone absorption spectrum in the UV: the Raman shifted wavelengths given in Table 4.2 are highlighted in this figure.

Ozone Raman DIAL

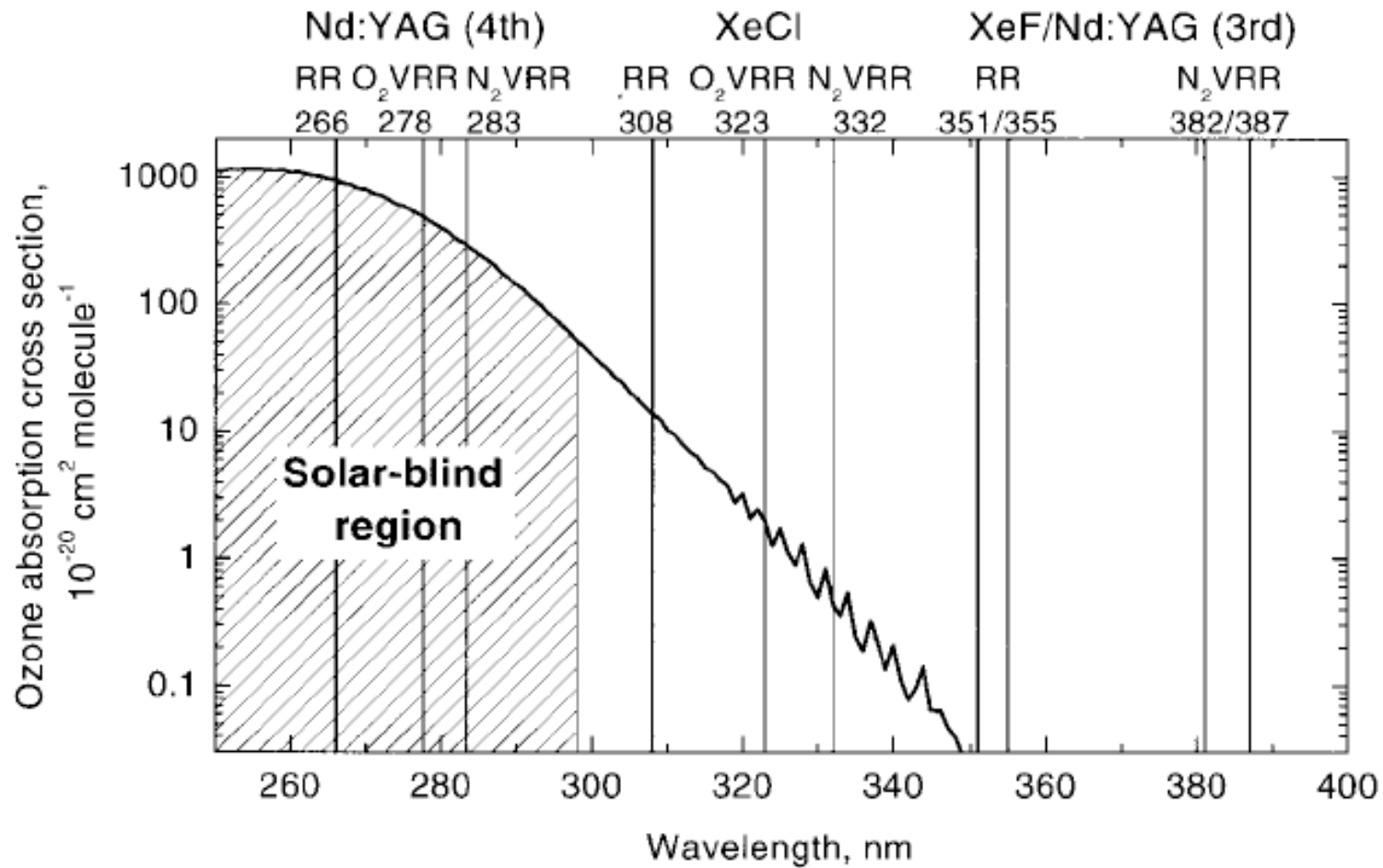
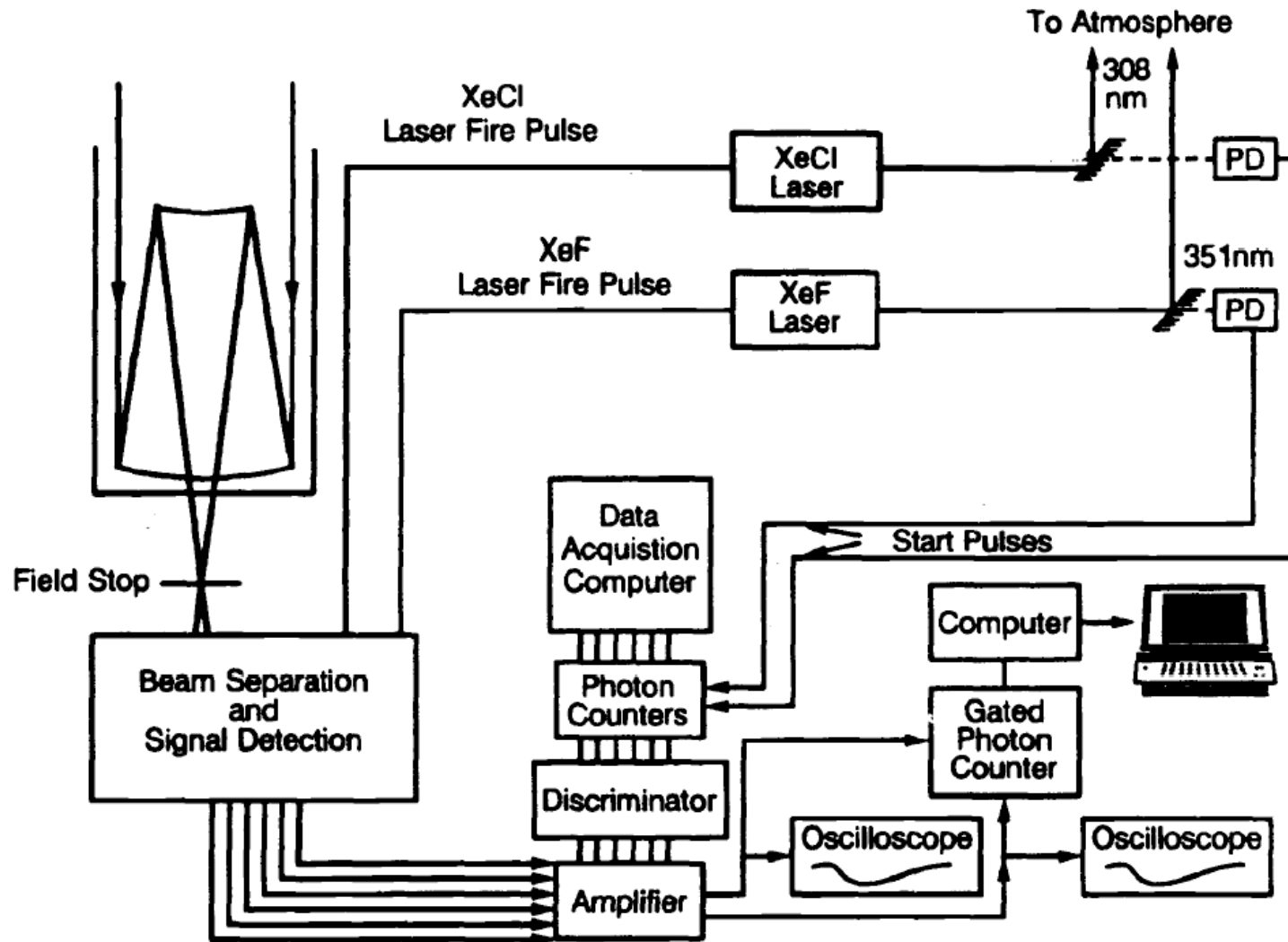
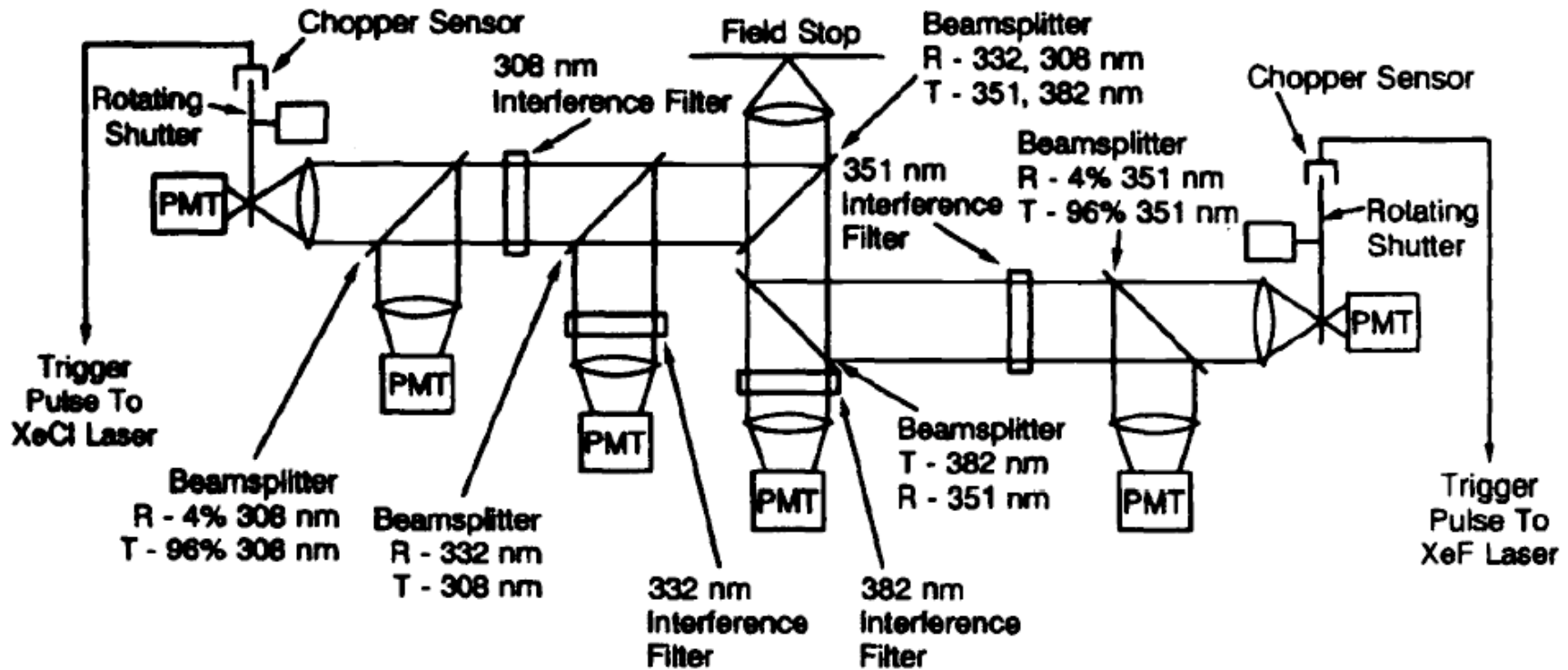


Fig. 9.5. Ozone absorption cross section and wavelengths applied in the ozone Raman DIAL technique for upper tropospheric and stratospheric measurements ($\lambda > 300 \text{ nm}$) and for boundary-layer measurements in the solar-blind region ($\lambda < 300 \text{ nm}$).

Ozone Raman DIAL



Ozone Raman DIAL



Raman DIAL Equation

□ For the normal DIAL channels at ON and OFF wavelengths,

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

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

Raman DIAL Equation Cont'd

□ For the Raman reference channels (e.g., from N₂ Raman scattering),

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

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

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

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

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.

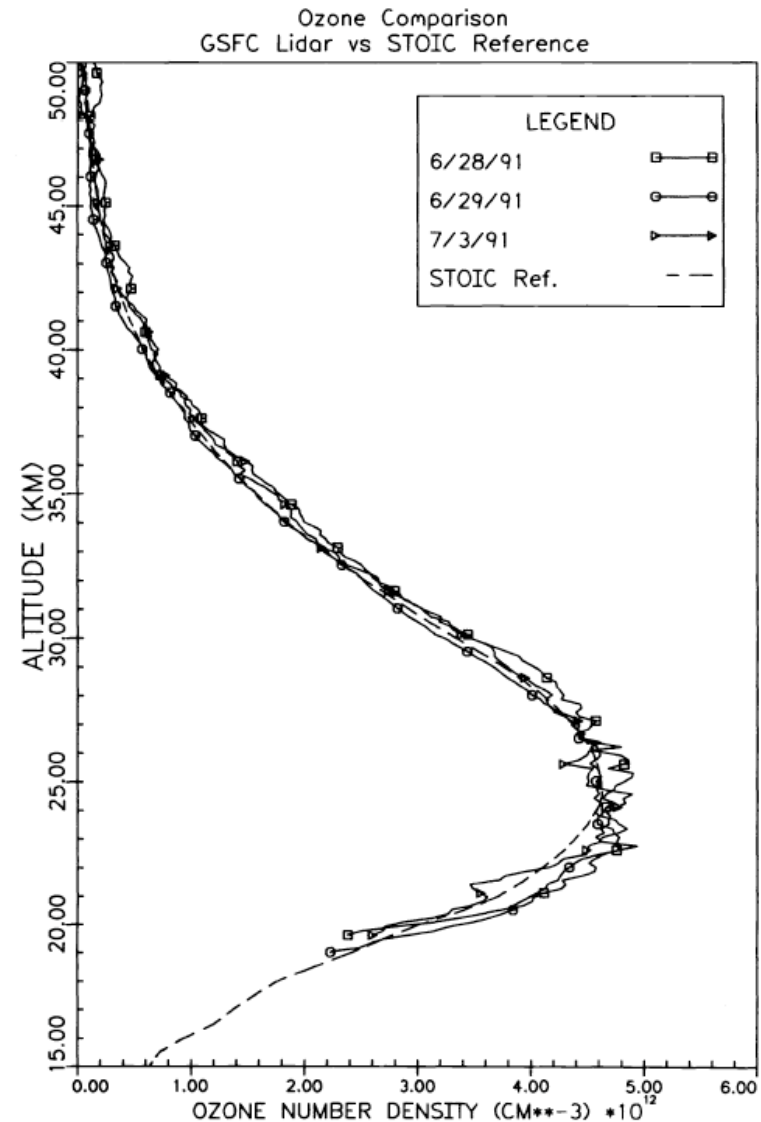


Fig. 6 GSFC lidar ozone profiles taken at JPL-TMO during June and July 1991 using the mechanical chopper.

Single-Laser RVR Raman DIAL

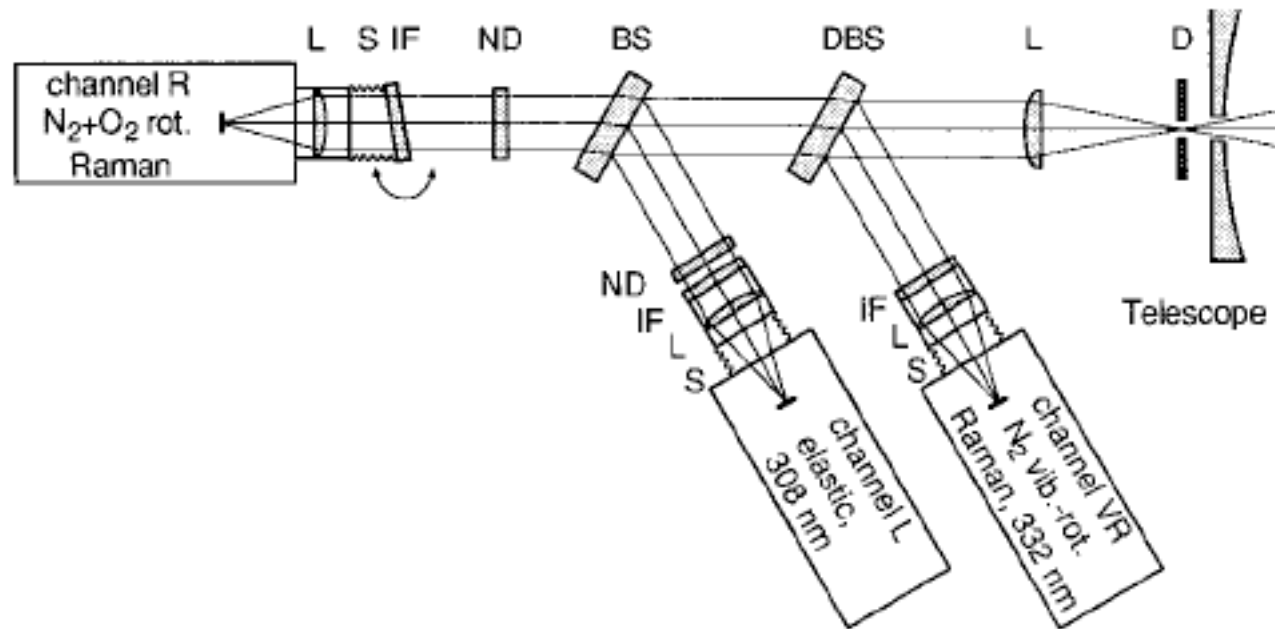


Fig. 3. RVR Raman DIAL receiver: D, diaphragm; L, lens; DBS, dichroic beam splitter; BS, beam splitter; ND, neutral-density filter; IF, interference filter; S, flexible shield. Atmospheric rotational Raman, elastic, and N_2 vibrational-rotational Raman backscattering signals are detected in channels R, L, and VR. The center wavelength of the channel-R interference filter can be tuned by rotating the filter around the vertical axis.

RVR Raman DIAL

Table 1. Parameters of the Conventional Raman DIAL and the RVR Raman DIAL's^a

Lidar	On-Resonance		Off-Resonance		$\Delta\lambda$ (nm)	$\Delta C_{\text{O}_3}^{\text{abs}}$ (10^{-24} m^2)
	Raman Wavelength (nm)	λ_{L} (nm)	Raman Wavelength (nm)	λ_{L} (nm)		
O ₂ RVR Raman DIAL	307	308	323	308	16	11.7
N ₂ RVR Raman DIAL	307	308	332	308	25	12.3
Raman DIAL	332	308	387	355	79	12.4

^a $\Delta\lambda$, maximum spectral separation of the signals used for the measurements. Differential ozone absorption cross sections ($\Delta C_{\text{O}_3}^{\text{abs}}$) are calculated from Ref. 13 ($T = 226 \text{ K}$). Wavelength values are rounded to full nanometers.

The RVR Raman DIAL measurement yields the numbers $N(\lambda, z)$ of lidar return photons from distance z at the rotational Raman wavelength λ_{R} of molecular oxygen and nitrogen and at the vibrational-rotational Raman wavelength λ_{VR} of O₂ or N₂, if light of the primary wavelength λ_{L} is transmitted. When absorption by particles and trace gases other than ozone is neglected, a condition satisfied for cirrus measurements in the free troposphere,³ the RVR Raman DIAL ozone molecule number density $n(z)$ in the single-scattering approximation is given by

RVR Raman DIAL

$$n(z) = RVRN - RVRM - RVRP, \quad (1)$$

where

$$RVRN = \frac{(d/dz)\ln[N(\lambda_{VR}, z)/N(\lambda_R, z)]}{C_{O_3}^{abs}(\lambda_R, T) - C_{O_3}^{abs}(\lambda_{VR}, T)},$$

$$RVRM = \frac{\alpha_{mol}^{sca}(\lambda_R, z) - \alpha_{mol}^{sca}(\lambda_{VR}, z)}{C_{O_3}^{abs}(\lambda_R, T) - C_{O_3}^{abs}(\lambda_{VR}, T)},$$

$$RVRP = \frac{\alpha_{par}^{sca}(\lambda_R, z) - \alpha_{par}^{sca}(\lambda_{VR}, z)}{C_{O_3}^{abs}(\lambda_R, T) - C_{O_3}^{abs}(\lambda_{VR}, T)}.$$

Here $C_{O_3}^{abs}(\lambda, T)$ is the ozone absorption cross section at temperature T , and $\alpha_{mol}^{sca}(\lambda, z)$ and $\alpha_{par}^{sca}(\lambda, z)$ are the Rayleigh extinction and the single-scattering particle extinction coefficient, respectively. Similar expressions, RDN , RDM , and RDP , have been derived for conventional Raman DIAL.⁴

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.