



Lecture 05. Fundamentals of Lidar Remote Sensing (3) - "Physical Processes in Lidar"

- ❑ Overview of physical processes in lidar
- ❑ Fundamentals behind physical processes
 - Atomic structure and energy levels
 - Molecular structure and energy levels
 - Energy bands of non-gas molecules
- ❑ Light interaction with objects
 - Elastic and in-elastic scattering
 - Absorption and differential absorption
 - Fluorescence and resonance fluorescence
 - Doppler effects and Boltzmann distribution
- ❑ Summary

General Lidar Equation in β and α

$$N_S(\lambda, R) = \left[\frac{P_L(\lambda_L) \Delta t}{hc/\lambda_L} \right] \left[\beta(\lambda, \lambda_L, \theta, R) \Delta R \right] \left(\frac{A}{R^2} \right) \cdot \exp \left[-\int_0^R \alpha(\lambda_L, r') dr' \right] \exp \left[-\int_0^R \alpha(\lambda, r') dr' \right] \left[\eta(\lambda, \lambda_L) G(R) \right] + N_B$$

β is the volume scatter coefficient (4.17)
 α is the extinction coefficient

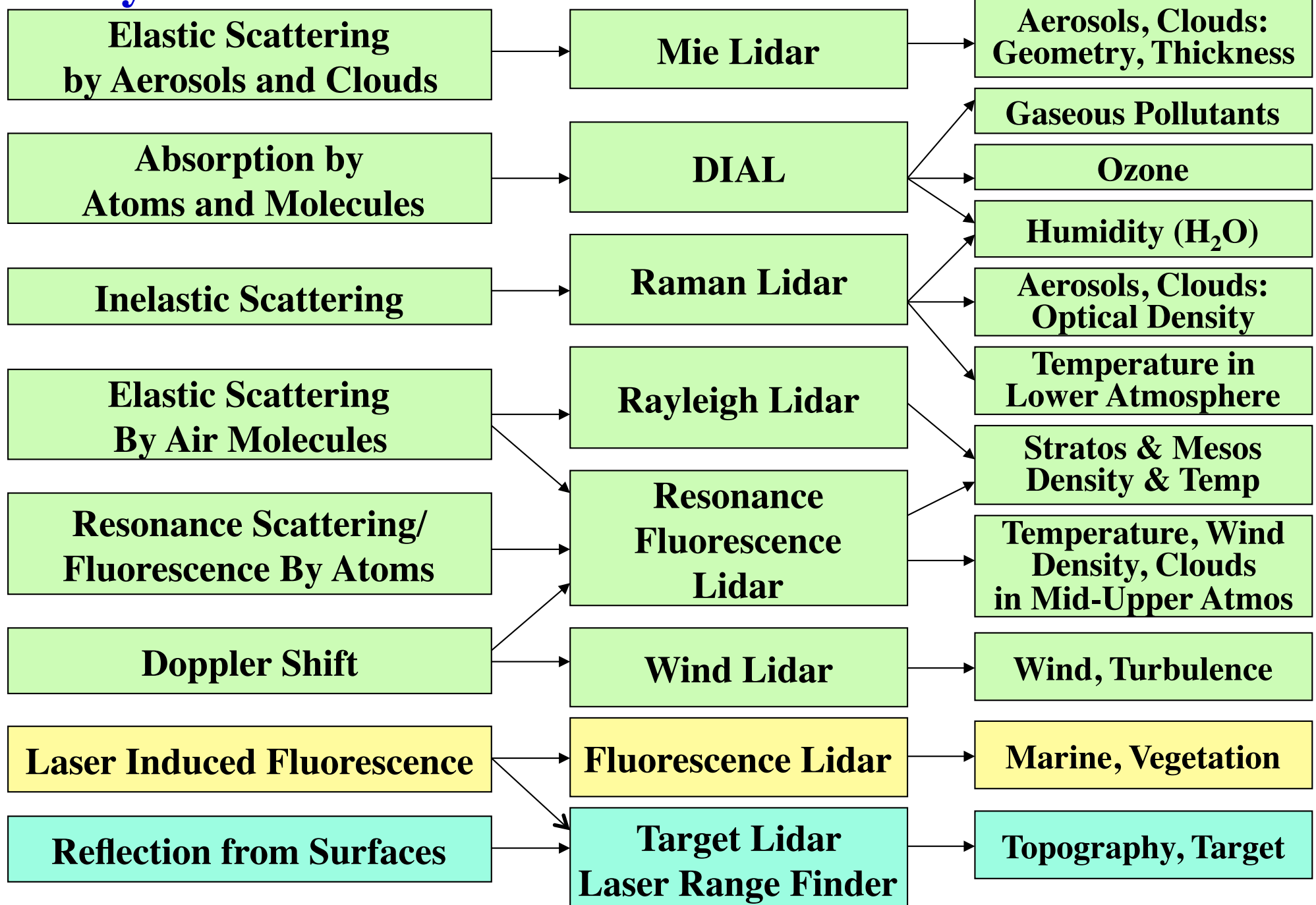
Volume scatter coefficient $\beta(\lambda, \lambda_L, R) = \sum_i \left[\frac{d\sigma_i(\lambda_L)}{d\Omega} n_i(R) p_i(\lambda) \right]$ (4.5)

Transmission $T(\lambda_L, R)T(\lambda, R) = \exp \left[-\left(\int_0^R \alpha(\lambda_L, r) dr + \int_0^R \alpha(\lambda, r) dr \right) \right]$ (4.18)

Physical Process

Device

Objective





Overview of Physical Processes in Lidar

❑ Interaction between light and objects

(1) Scattering (instantaneous elastic & inelastic):

Mie, Rayleigh, and Raman scattering

(2) Absorption and differential absorption

(3) Resonance fluorescence

(4) Laser induced fluorescence

(5) Doppler shift and Doppler broadening

(6) Boltzmann distribution

(7) Reflection from target or surface

(8) Multiple scattering and polarization in scattering

❑ Light propagation in the atmosphere or medium:

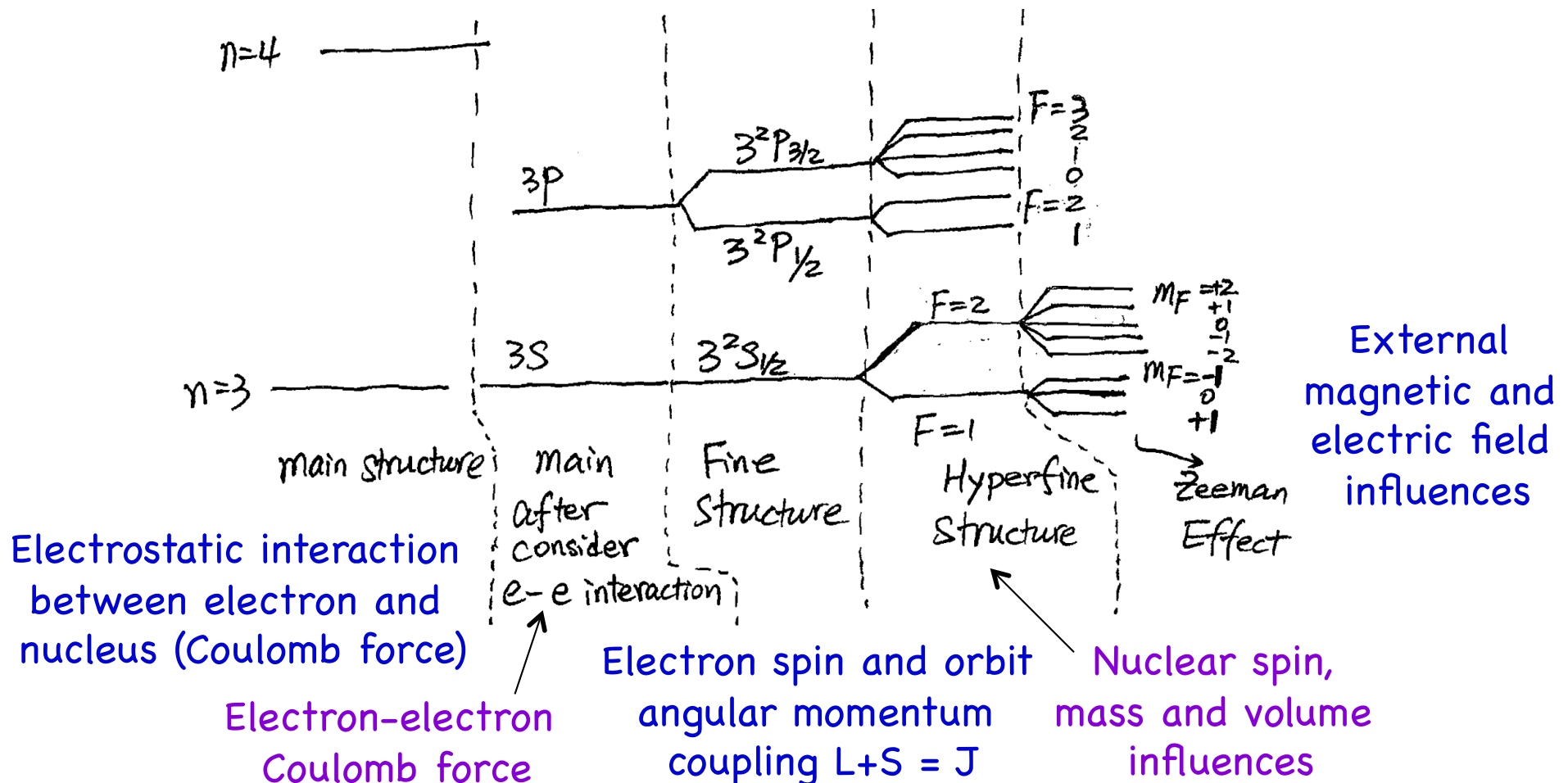
Light transmission vs. light extinction (attenuation)

Extinction (attenuation) = Absorption + Scattering (Total)

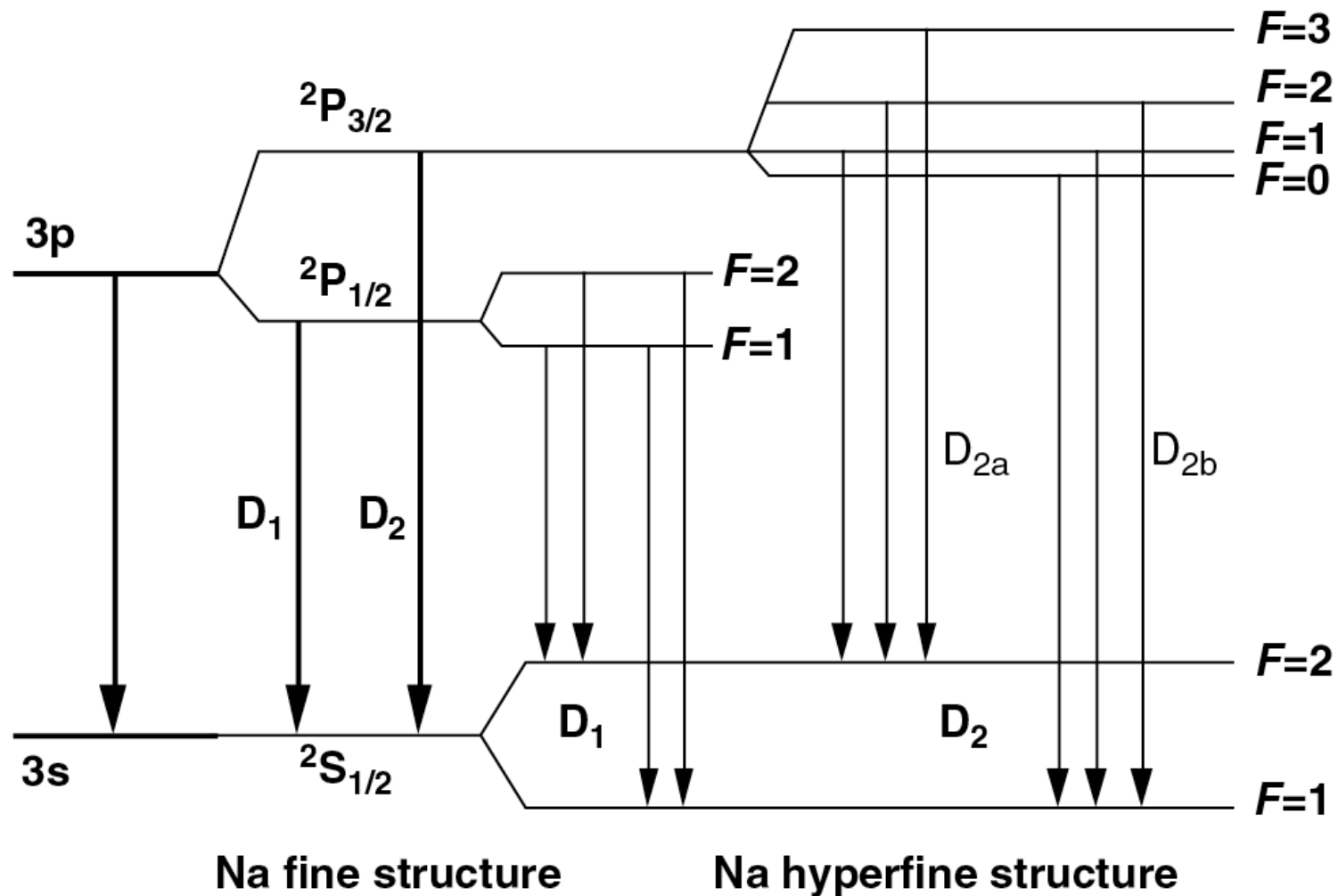
Atomic Structure and Energy Levels

□ Interactions inside and outside an atom determine the atomic energy levels that are usually concrete levels. A class on "Fundamentals of Spectroscopy for Optical Remote Sensing" can be found at

<http://cires.colorado.edu/science/groups/chu/classes/Spectroscopy2013/>



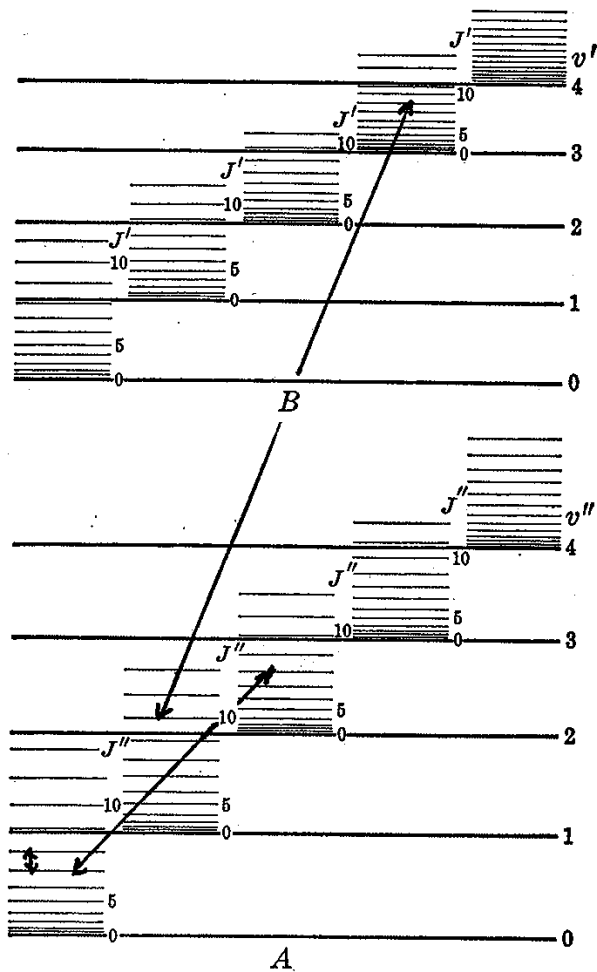
Example: Na D_2 Lines



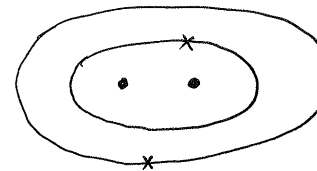


Molecular Structure and Energy Levels

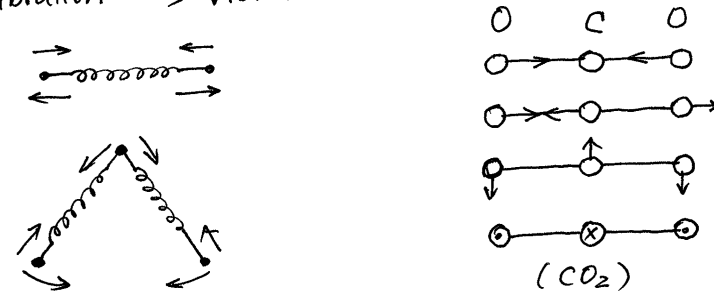
□ A molecule contains more interactions than an atom because of the interaction among atoms and motions within a molecule.



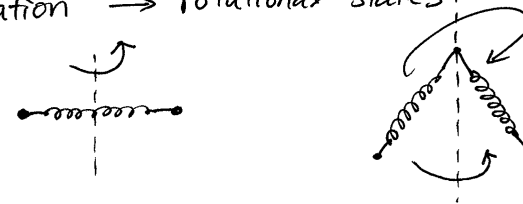
(1) Electron motion (mainly outer electrons) → electronic state



(2) Nucleus vibration → Vibrational states



(3) Nucleus rotation → rotational states

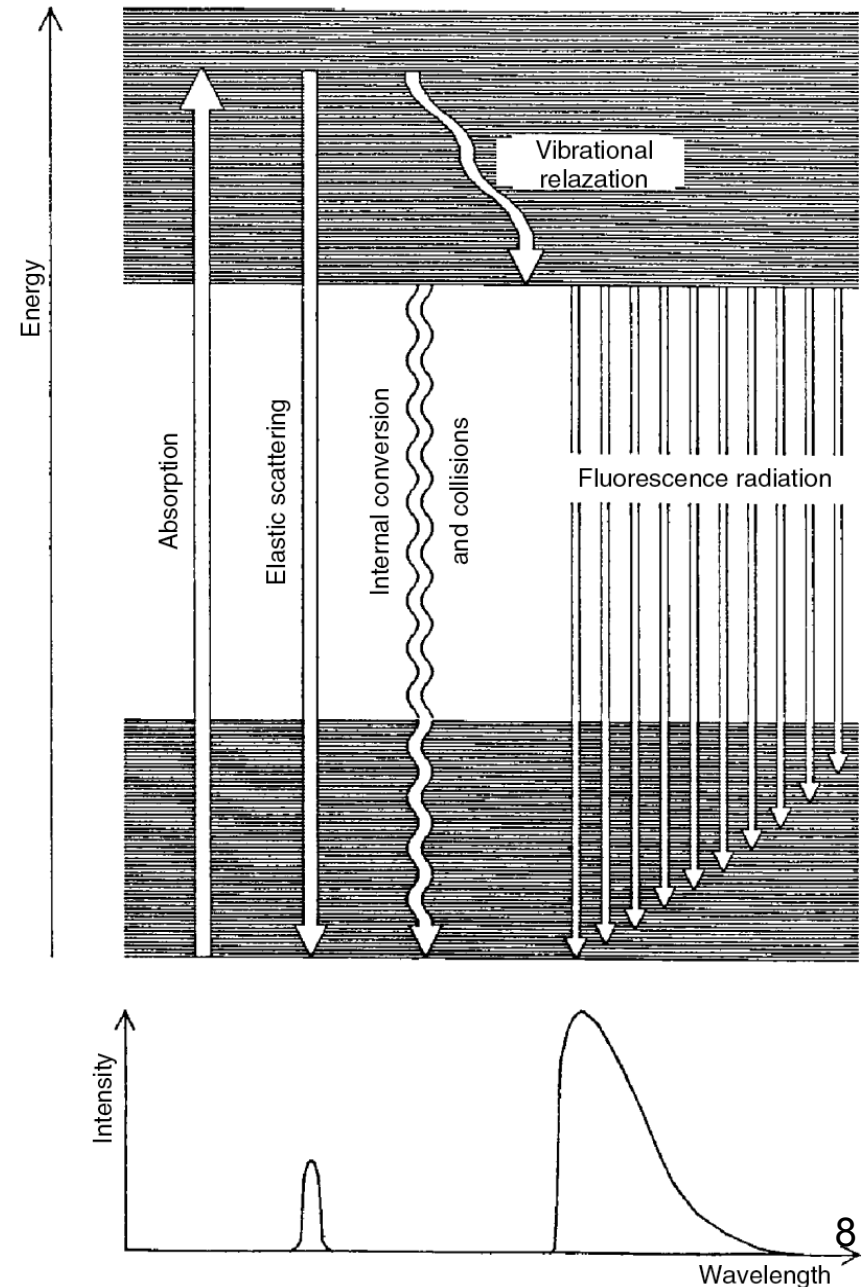


(4) Electron spin, nuclear spin → fine and hyperfine structure of the electronic states

(5) Translation of the entire molecule → continuum energy

Energy Bands of Non-Gas Molecules

- ❑ In contrast to free atoms and molecules, solids and liquids exhibit broad absorption and emission spectra because of the strong intermolecular interactions.
- ❑ Following the laser excitation, there is a very fast (ps) radiationless relaxation down to the lowest sub-level of the excited state, where the molecules remain for a typical excited-state fluorescence lifetime.
- ❑ The decay then occurs to different sub-levels of the ground state giving rise to a distribution of fluorescence light, which reflect the lower-state level distribution.





Elastic and Inelastic Scattering

□ Elastic scattering:

scattering with no apparent change of wavelength

Sum of elastic scattering from atmospheric molecules and elastic scattering from aerosol particles and cloud droplets

Rayleigh scattering and Mie scattering

(There are confusions in definitions of these scatterings.)

□ Inelastic scattering:

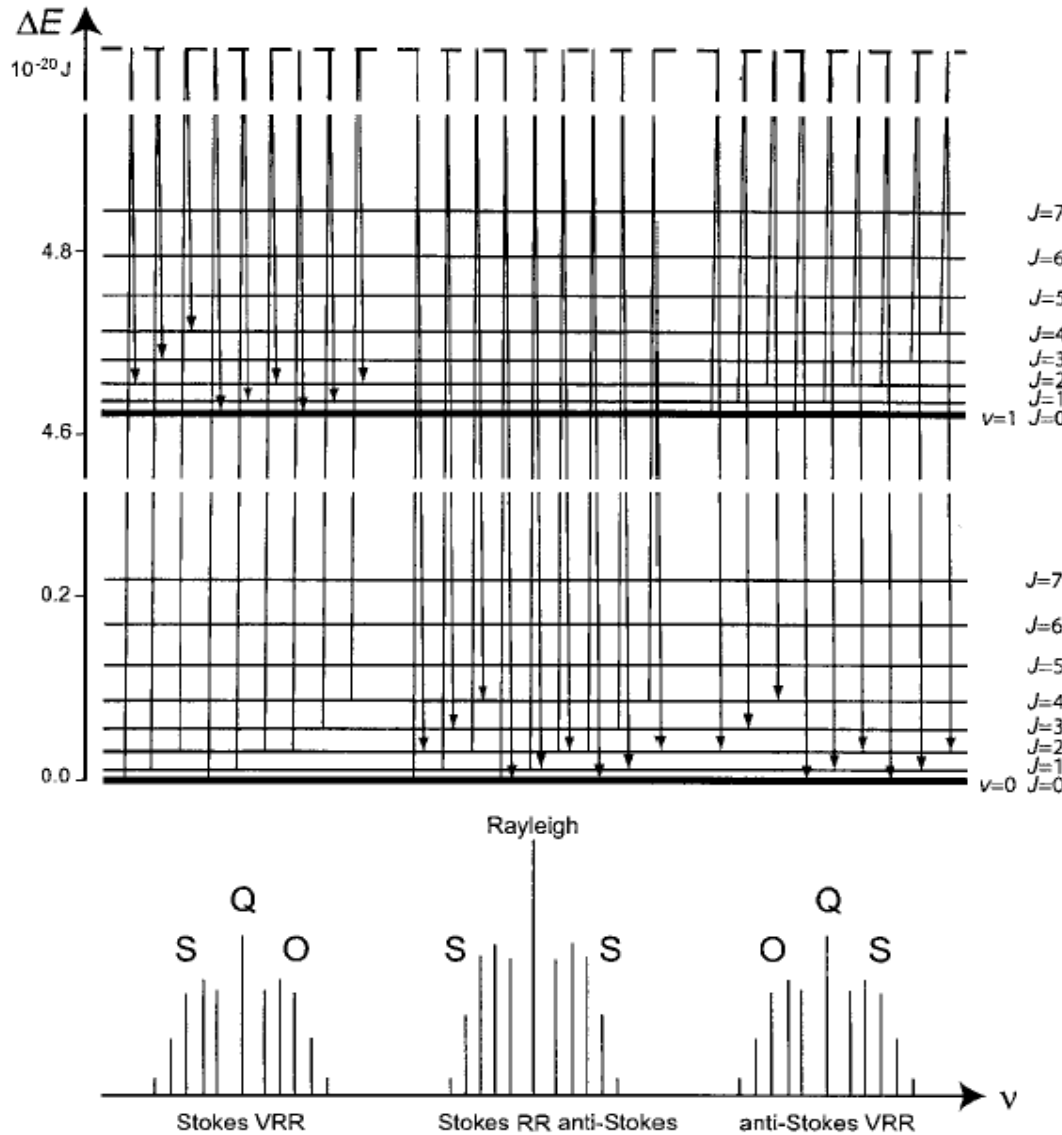
scattering with apparent change of wavelength

Raman scattering

Pure rotational Raman and vibration-rotational Raman



Definitions for Different Scatterings



Virtual Energy Levels

Real Energy Levels

Real Energy Levels

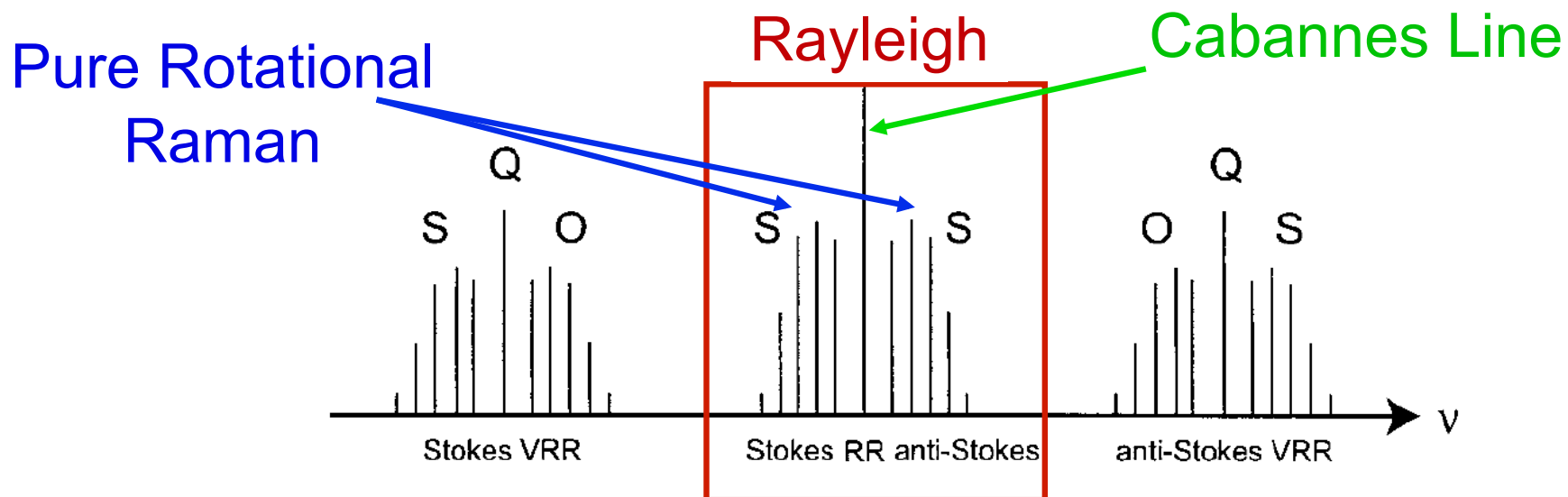
$$\Delta v = 0, \quad \Delta J = 2$$

$$\Delta v = \pm 1, \quad \Delta J = 0, \pm 2$$

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

Rayleigh (Molecular) Scattering

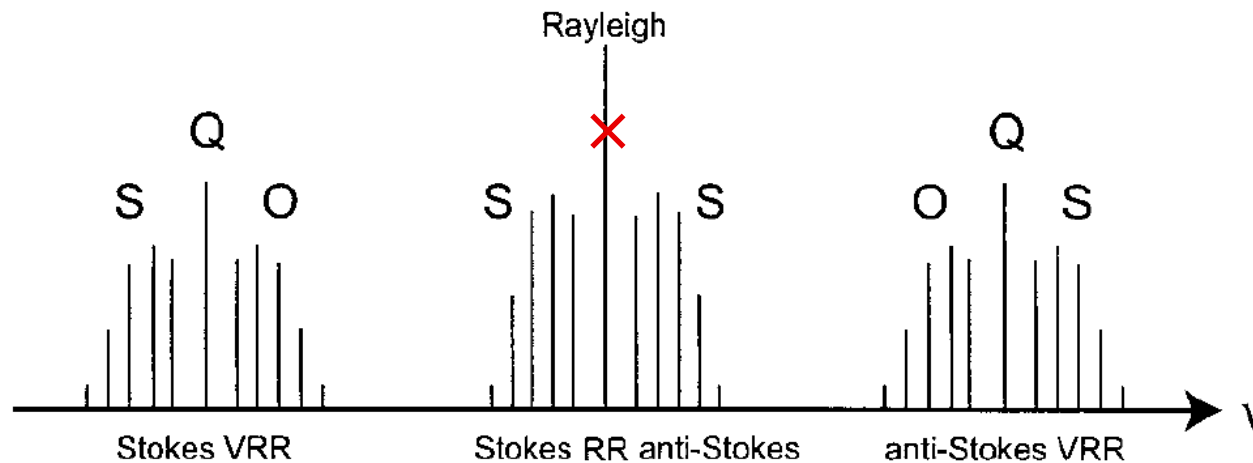
- Rayleigh scattering is referred to the elastic scattering from atmospheric molecules (particle size is much smaller than the wavelength), i.e., scattering with no apparent change of wavelength, although still undergoing Doppler broadening and Doppler shift.
- However, depending on the resolution of detection, Rayleigh scattering can consist of the Cabannes scattering (really elastic scattering from molecules) and pure rotational Raman scattering.





Raman Scattering

□ Raman scattering is an inelastic scattering with rotational quantum state or vibration-rotational quantum state change as the result of scattering.



□ Separation between Cabannes line and the nearest pure rotational Raman (O_2 and N_2) is about $10 \text{ cm}^{-1} = 300 \text{ GHz}$.

□ For vibration-rotational Raman, Q branch is not a single line, but consists of many lines with very small separations, $< 0.1 \text{ cm}^{-1} = 3 \text{ GHz}$, due to the different splitting of the rotational levels in the lower and upper vibrational levels.

Mie (Aerosol/Cloud) Scattering

- ❑ Strictly speaking, Mie scattering is an elastic scattering from spherical particles [Mie, 1908], which includes the solution of Rayleigh scattering.
- ❑ However, in lidar field, Mie scattering is referred to the elastic scattering from spherical particles whose size is comparable to or larger than the wavelength.
- ❑ Furthermore, Mie scattering is generalized to elastic scattering from overall aerosol particles and cloud droplets, i.e., including non-spherical particles.
- ❑ To precisely calculate the scattering from non-spherical particles, Mie scattering theory has to be replaced by non-spherical particle scattering theories. This is a complicated issue in the elastic lidar field.

Rayleigh Backscatter Coefficient

- Precise equations should be obtained from Rayleigh theory and experimental measurements of parameters.
- In lidar field, a common practice is to use the equation

$$\beta_{Rayleigh}(\lambda, z, \theta = \pi) = 2.938 \times 10^{-32} \frac{P(z)}{T(z)} \cdot \frac{1}{\lambda^{4.0117}} \left(m^{-1} sr^{-1} \right) \quad (5.1)$$

where P is the atmosphere pressure in mbar and T is the temperature in Kelvin at altitude z , λ is the wavelength in meter, and $\beta_{Rayleigh}$ is the backscatter coefficient (angular).

- Total scatter coefficient β_T has relationship:

$$\beta(\theta) = \frac{\beta_T}{4\pi} P(\theta) = \frac{\beta_T}{4\pi} \times 0.7629 \times (1 + 0.9324 \cos^2 \theta) \quad (5.2)$$



Rayleigh Backscatter Cross Section

□ It is also common in lidar field to calculate the Rayleigh backscatter cross section using the following equation

$$\frac{d\sigma_m(\lambda)}{d\Omega} = 5.45 \cdot \left(\frac{550}{\lambda}\right)^4 \times 10^{-32} \left(m^2 sr^{-1}\right) \quad (5.3)$$

where λ is the wavelength in nm.

- For K lidar, $\lambda = 770$ nm, $\Rightarrow 1.42 \times 10^{-32} m^2 sr^{-1}$
- For Na lidar, $\lambda = 589$ nm, $\Rightarrow 4.14 \times 10^{-32} m^2 sr^{-1}$
- For Fe lidar, $\lambda = 372$ nm, $\Rightarrow 2.60 \times 10^{-31} m^2 sr^{-1}$
- For Rayleigh lidar, $\lambda = 532$ nm, $\Rightarrow 6.22 \times 10^{-32} m^2 sr^{-1}$



Scattering Form of Lidar Equation

□ Rayleigh, Mie, and Raman scattering processes are instantaneous scattering processes, so there are no finite relaxation effects involved, but infinitely short duration.

□ For **Rayleigh and Mie scattering**, there is no frequency shift when the atmospheric particles are at rest. The lidar equation is written as

$$N_S(\lambda, R) = \left(\frac{P_L(\lambda)\Delta t}{hc/\lambda} \right) (\beta(\lambda, R)\Delta R) \left(\frac{A}{R^2} \right) T^2(\lambda, R) (\eta(\lambda)G(R)) + N_B \quad (5.4)$$

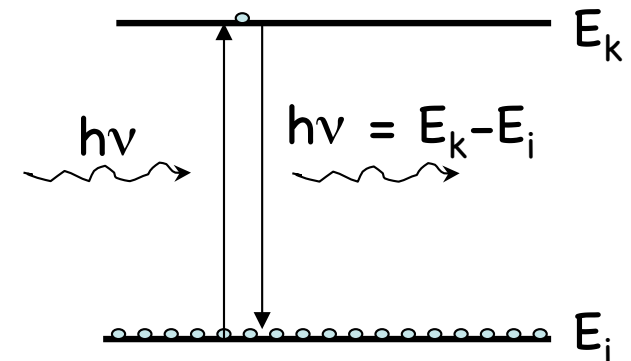
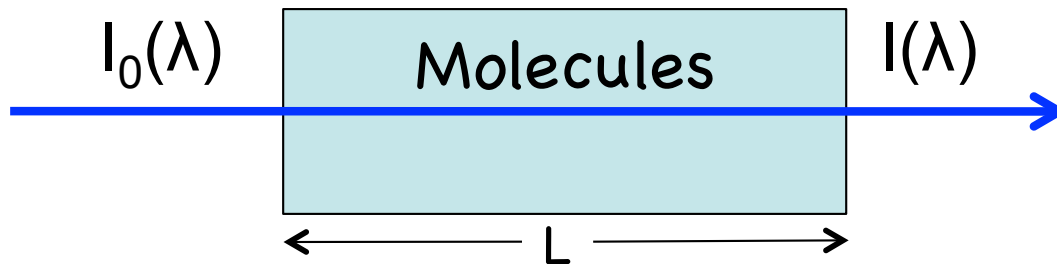
□ For **Raman scattering**, there is a large frequency shift. Raman lidar equation may be written as (5.5)

$$N_S(\lambda, R) = \left(\frac{P_L(\lambda_L)\Delta t}{hc/\lambda_L} \right) (\beta(\lambda, \lambda_L, R)\Delta R) \left(\frac{A}{R^2} \right) (T(\lambda_L, R)T(\lambda, R)) (\eta(\lambda, \lambda_L)G(R)) + N_B$$

$$\lambda \neq \lambda_L, \quad p_i(\lambda) \neq 1, \quad p_i(\lambda) < 1$$

$$T(\lambda, R) = \exp \left[- \int_0^R \alpha(\lambda, r) dr \right]$$

Molecular Absorption



➤ The intensity change dI of a light propagating through an absorbing sample is determined by the absorption coefficient $\alpha_{\text{mol,abs}}$ in the following manner:

$$dI(\lambda) = -I(\lambda)\alpha(\lambda)dz = -I\sigma_{ik}(\lambda)(N_i - N_k)dz \quad (5.6)$$

$\alpha(\lambda) = \sigma_{ik}(\lambda)(N_i - N_k)$ is absorption coefficient caused by transition $E_i \rightarrow E_k$. Here, σ_{ik} is the absorption cross section, N_i and N_k are the populations on the energy levels of E_i and E_k , respectively.

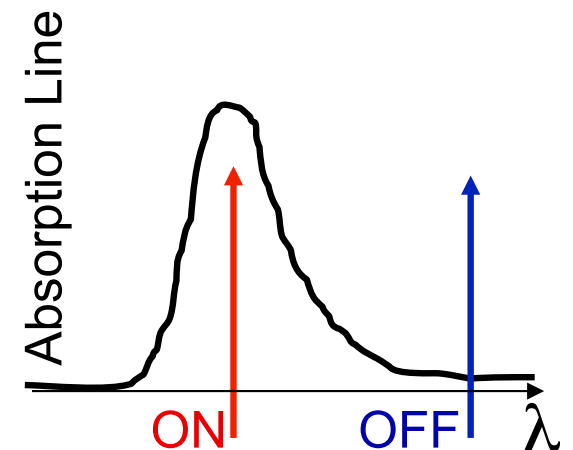
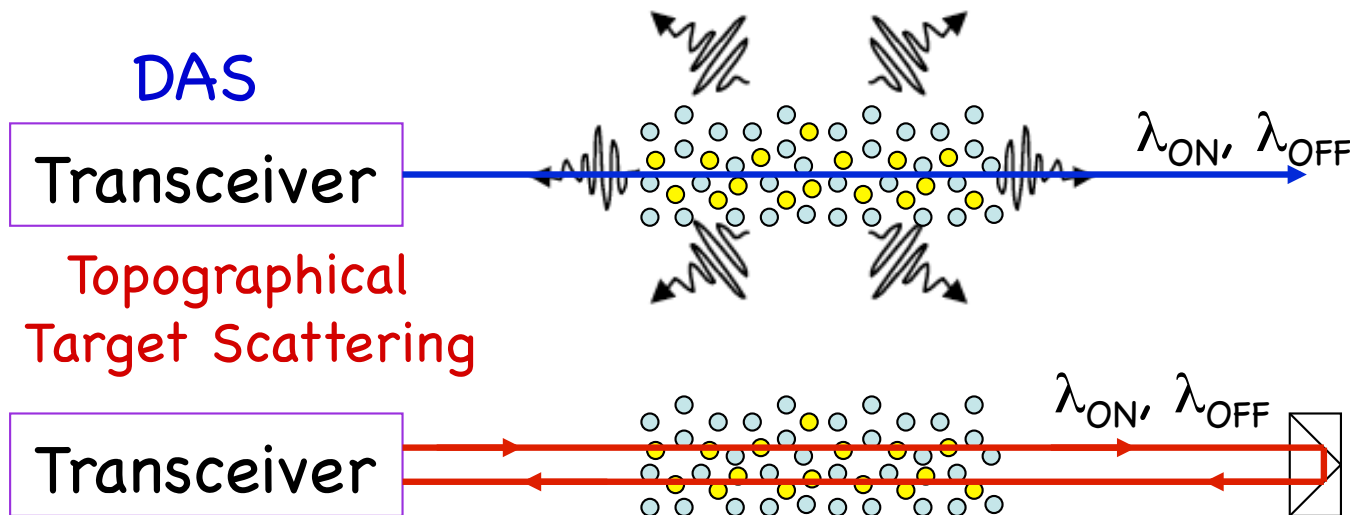
➤ If $\Delta N = N_i - N_k$ is independent of the light intensity I , the absorbed intensity dI is proportional to the incident intensity I (linear absorption). Solving the above equation, we obtain

$$I(\lambda, z) = I_0 \exp \left[-\int_0^z \sigma_{ik}(\lambda)(N_i - N_k) dz \right] \quad (5.7)$$

$$\Rightarrow I(\lambda, z) = I_0 e^{-\sigma(\lambda)(N_i - N_k)L} = I_0 e^{-\alpha(\lambda)L} \quad \text{-- Lambert-Beer's Law} \quad (5.8) \quad 17$$

Differential Absorption

- For the molecular species to be detected by DIAL, usually they have strong absorption, however, nearly none resonance fluorescence due to strong relaxation processes other than fluorescence (e.g., frequent collisions with surrounding atmosphere molecules can make molecules decay from excited states to ground state without giving fluorescence).
- Thus, in the lidar equation for DIAL, the influence of molecular species is in the extinction (atmosphere transmission) part, not in the backscatter part. In other words, the molecular absorption contributes to the extinction of light when incident light and scattered light propagate through atmosphere, while the return signals are from the scattering of laser light by air molecules and aerosols.





Differential Absorption/Scattering Form of Lidar Equation

- For the laser with wavelength λ_{on} on the molecular absorption line

$$N_S(\lambda_{on}, R) = N_L(\lambda_{on}) [\beta_{sca}(\lambda_{on}, R) \Delta R] \left(\frac{A}{R^2} \right) \exp \left[-2 \int_0^z \bar{\alpha}(\lambda_{on}, r') dr' \right] \times \exp \left[-2 \int_0^z \sigma_{abs}(\lambda_{on}, r') n_c(r') dr' \right] [\eta(\lambda_{on}) G(R)] + N_B \quad (5.9)$$

- For the laser with wavelength λ_{off} off the molecular absorption line

$$N_S(\lambda_{off}, R) = N_L(\lambda_{off}) [\beta_{sca}(\lambda_{off}, R) \Delta R] \left(\frac{A}{R^2} \right) \exp \left[-2 \int_0^z \bar{\alpha}(\lambda_{off}, r') dr' \right] \times \exp \left[-2 \int_0^z \sigma_{abs}(\lambda_{off}, r') n_c(r') dr' \right] [\eta(\lambda_{off}) G(R)] + N_B \quad (5.10)$$

- Differential absorption cross-section

$$\Delta \sigma_{abs}(R) = \sigma_{abs}(\lambda_{ON}, R) - \sigma_{abs}(\lambda_{OFF}, R) \quad (5.11)$$

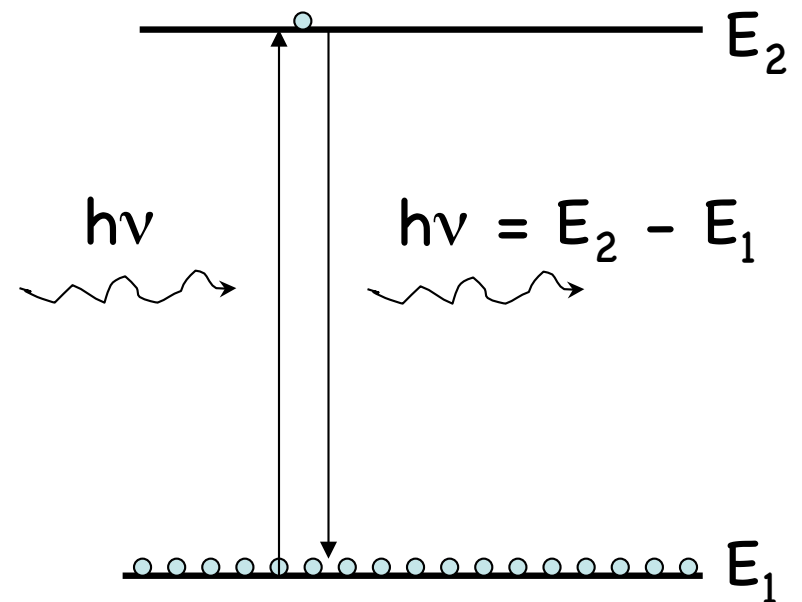


Resonance Fluorescence

□ In the middle and upper atmosphere, there exist some metal atoms and atomic ions. They have large absorption cross section, and quenching is not a problem in that region. Therefore, laser tuned to the resonance frequency of the absorption lines can excite resonance fluorescence from these atoms and ions.

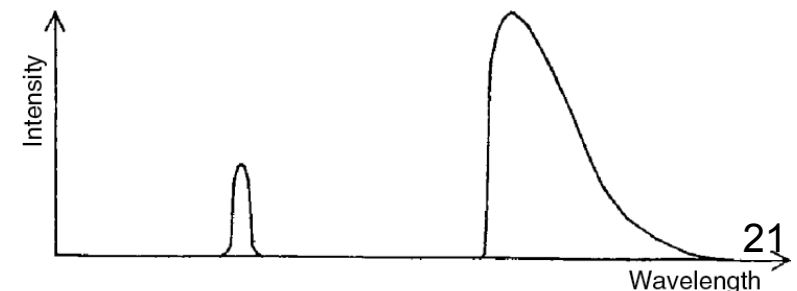
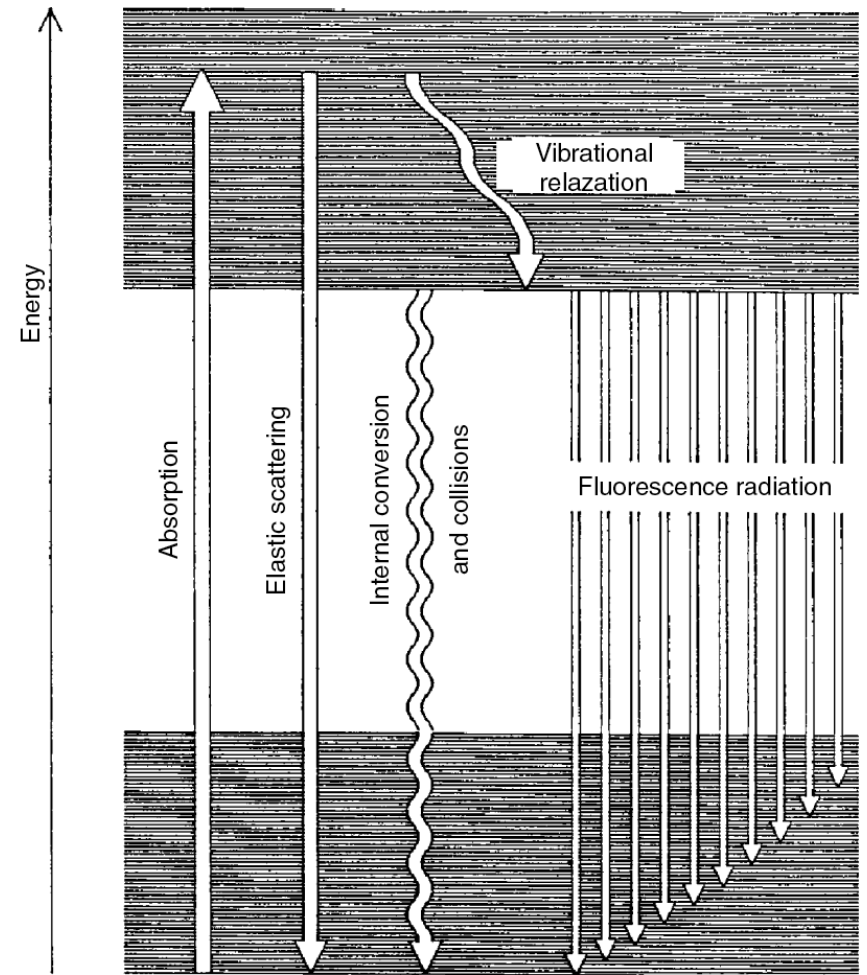
□ Resonance fluorescence is a two-step process: absorption first, and then spontaneous emission. Therefore, there is finite time delay between these two steps - radiative lifetime.

□ Due to frequent collisions it is hard to obtain resonance fluorescence in lower atmosphere.



Laser Induced Fluorescence

- ❑ In contrast to free atoms and molecules, solids and liquids exhibit broad absorption and emission spectra because of the strong intermolecular interactions.
- ❑ A fixed frequency laser can be used for the excitation due to the broad absorption.
- ❑ Following the excitation, there is a very fast (ps) radiationless relaxation down to the lowest sub-level of the excited state, where the molecules remain for a typical excited-state fluorescence lifetime.
- ❑ The decay then occurs to different sub-levels of the ground state giving rise to a distribution of fluorescence light, which reflect the lower-state level distribution.
- ❑ Fixing the excitation wavelength, we can obtain fluorescence spectra. While fixing the detection channel and varying the excitation wavelength, an excitation spectrum can be recorded.





Fluorescence Form of Lidar Equation

❑ Resonance fluorescence and laser-induced-fluorescence are NOT instantaneous processes, but have delays due to the radiative lifetime of the excited states.

❑ The lidar equation in fluorescence form is given by (5.12)

$$N_S(\lambda, R) = \left(\frac{P_L(\lambda) \Delta t}{hc/\lambda} \right) \left(\sigma_{eff}(\lambda, R) n_c(z) R_B(\lambda) \Delta R \right) \left(\frac{A}{4\pi R^2} \right) \left(T_a^2(\lambda, R) T_c^2(\lambda, R) \right) (\eta(\lambda) G(R)) + N_B$$

❑ Here, $T_c(R)$ is the transmission caused by the constituent absorption.

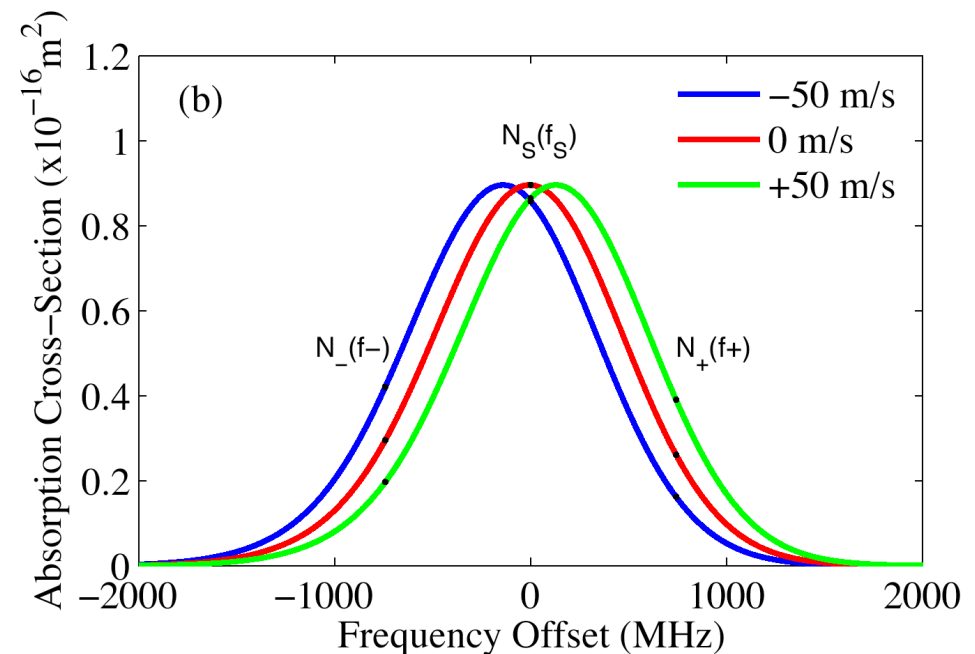
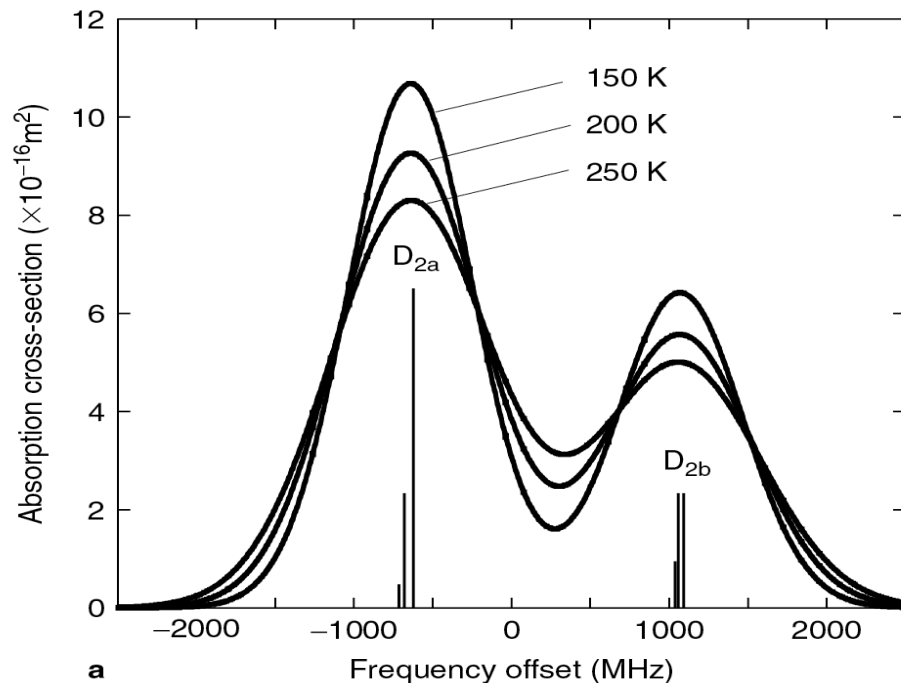
$$T_c(R) = \exp\left(-\int_{R_{bottom}}^R \sigma_{eff}(\lambda, r') n_c(r') dr'\right) = \exp\left(-\int_{R_{bottom}}^R \alpha_c(\lambda, r') dr'\right) \quad (5.13)$$

❑ Here, $\alpha(\lambda, R)$ is the extinction coefficient caused by the absorption.

$$\alpha_c(\lambda, R) = \sigma_{eff}(\lambda, R) n_c(R) \quad (5.14)$$

Doppler Shift and Broadening

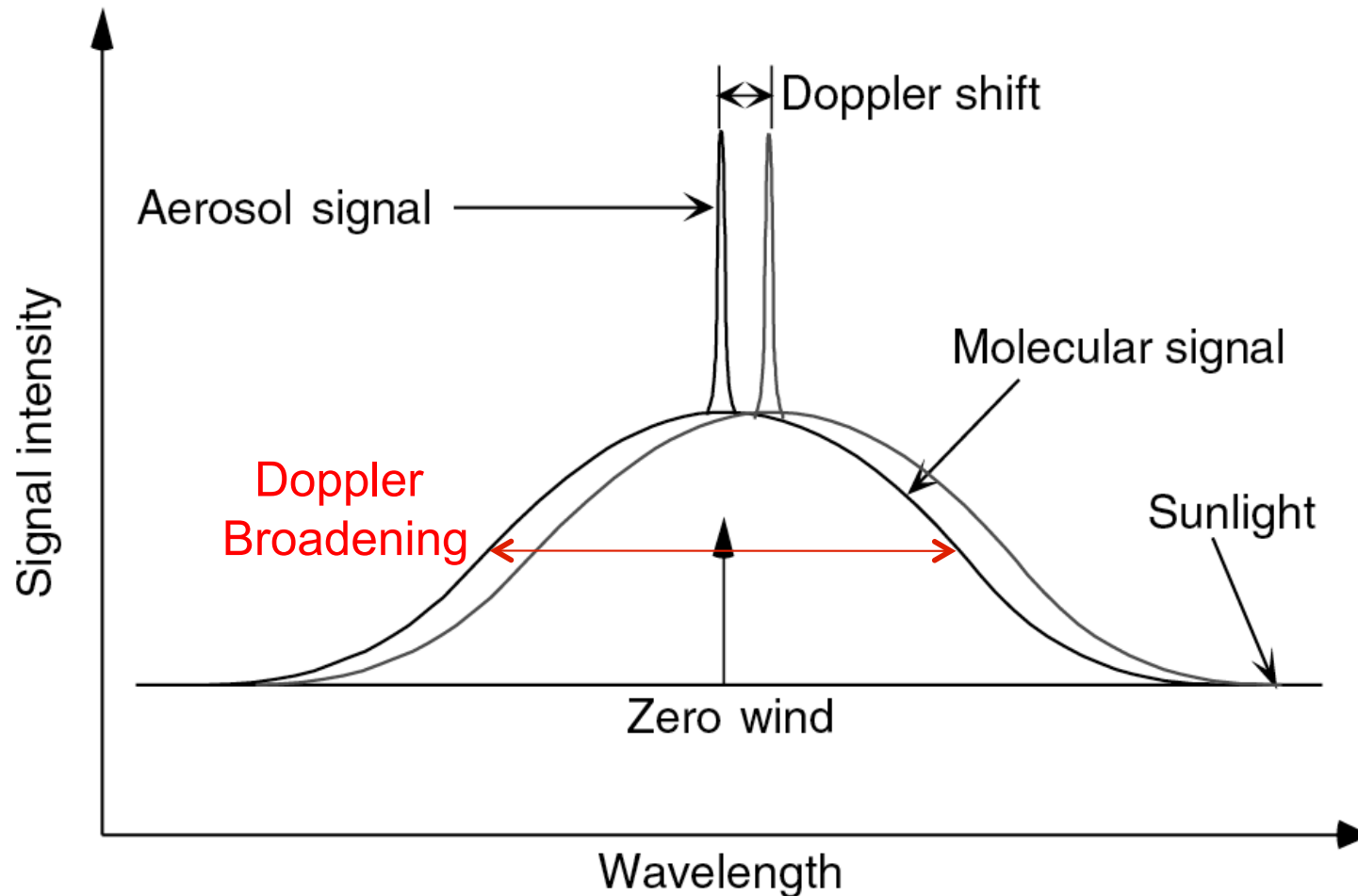
□ **Doppler Effects** – Doppler linewidth broadening and Doppler frequency shift are temperature-dependent and wind-dependent, respectively (applying to both Na, K, Fe resonance fluorescence and molecular scattering)



$$\sigma_{rms} = \frac{\omega_0}{c} \sqrt{\frac{k_B T}{M}} = \frac{1}{\lambda_0} \sqrt{\frac{k_B T}{M}} \quad (5.15)$$

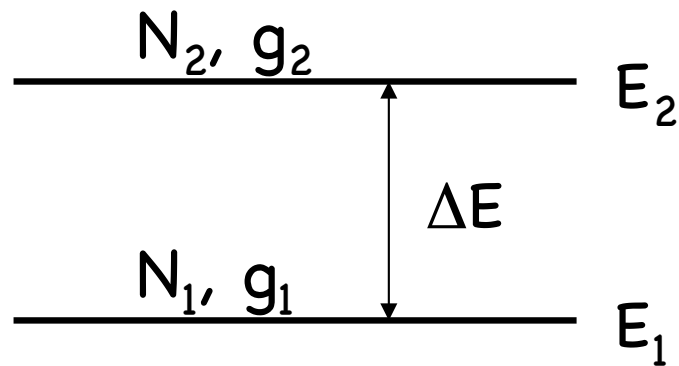
$$\Delta\omega = \omega - \omega_0 = -\vec{k} \cdot \vec{v} = -\omega_0 \frac{v \cos\theta}{c} \quad (5.16)$$

Doppler Shift and Broadening in Atmospheric Scattering Signals



Boltzmann Distribution

□ Maxwell-Boltzmann distribution is the law of particle population distribution according to energy levels (under thermodynamic equilibrium)



$$\frac{N_k}{N} = \frac{g_k \exp(-E_k / k_B T)}{\sum_i g_i \exp(-E_i / k_B T)} \quad (5.17)$$

$$\frac{N_2}{N_1} = \frac{g_2}{g_1} \exp\left\{-\frac{(E_2 - E_1)}{k_B T}\right\} \quad (5.18)$$

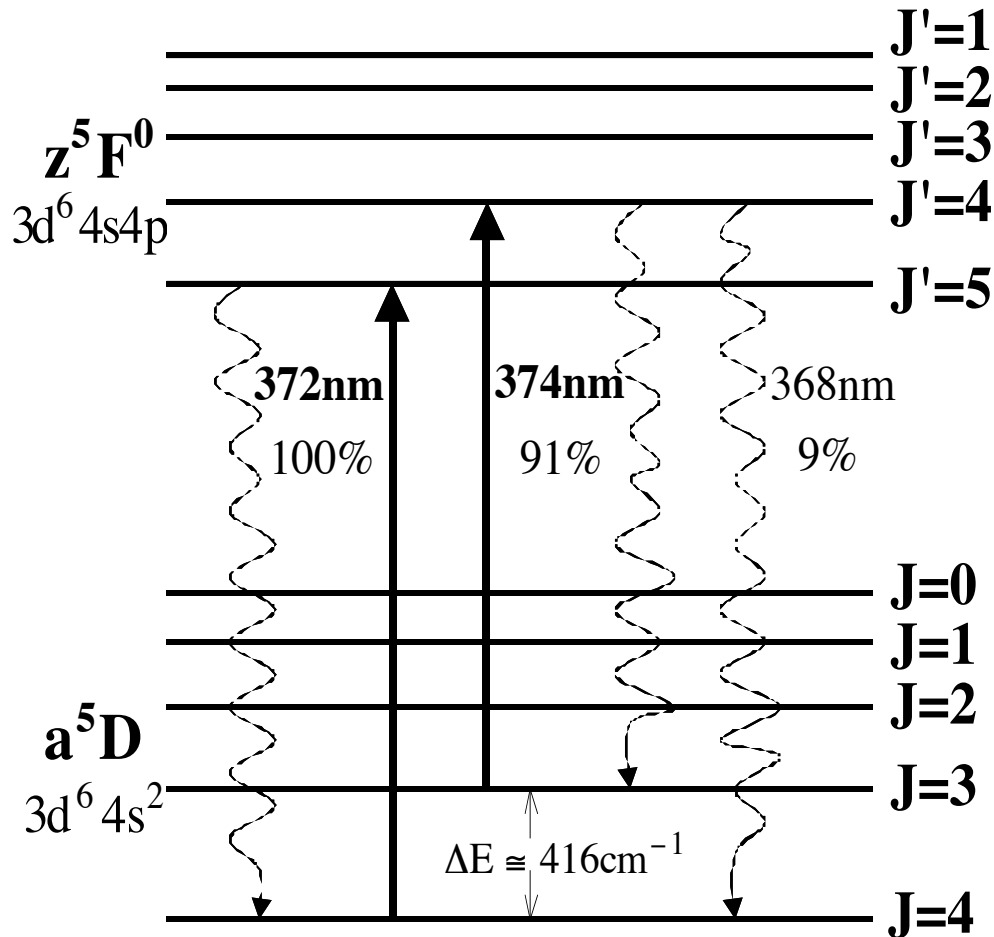
$$T = \frac{\Delta E / k_B}{\ln\left(\frac{g_2 \cdot N_1}{g_1 \cdot N_2}\right)} \quad (5.19)$$

N_1 and N_2 - particle populations on energy levels E_1 and E_2
 g_1 and g_2 - degeneracy for energy levels E_1 and E_2 , $\Delta E = E_2 - E_1$
 k_B - Boltzmann constant, T - Temperature, N - total population

Population Ratio \Rightarrow Temperature



Fe Boltzmann Technique



Atomic Fe Energy Level

[Gelbwachs, 1994; Chu et al., 2002]

Example: Fe Boltzmann

$$\frac{N(J=4)}{N(J=3)} = \frac{g_1}{g_2} \exp\{\Delta E/k_B T\}$$

$$g_1 = 2 * 4 + 1 = 9$$

$$g_2 = 2 * 3 + 1 = 7$$

$$\Delta E = 416(\text{cm}^{-1})$$

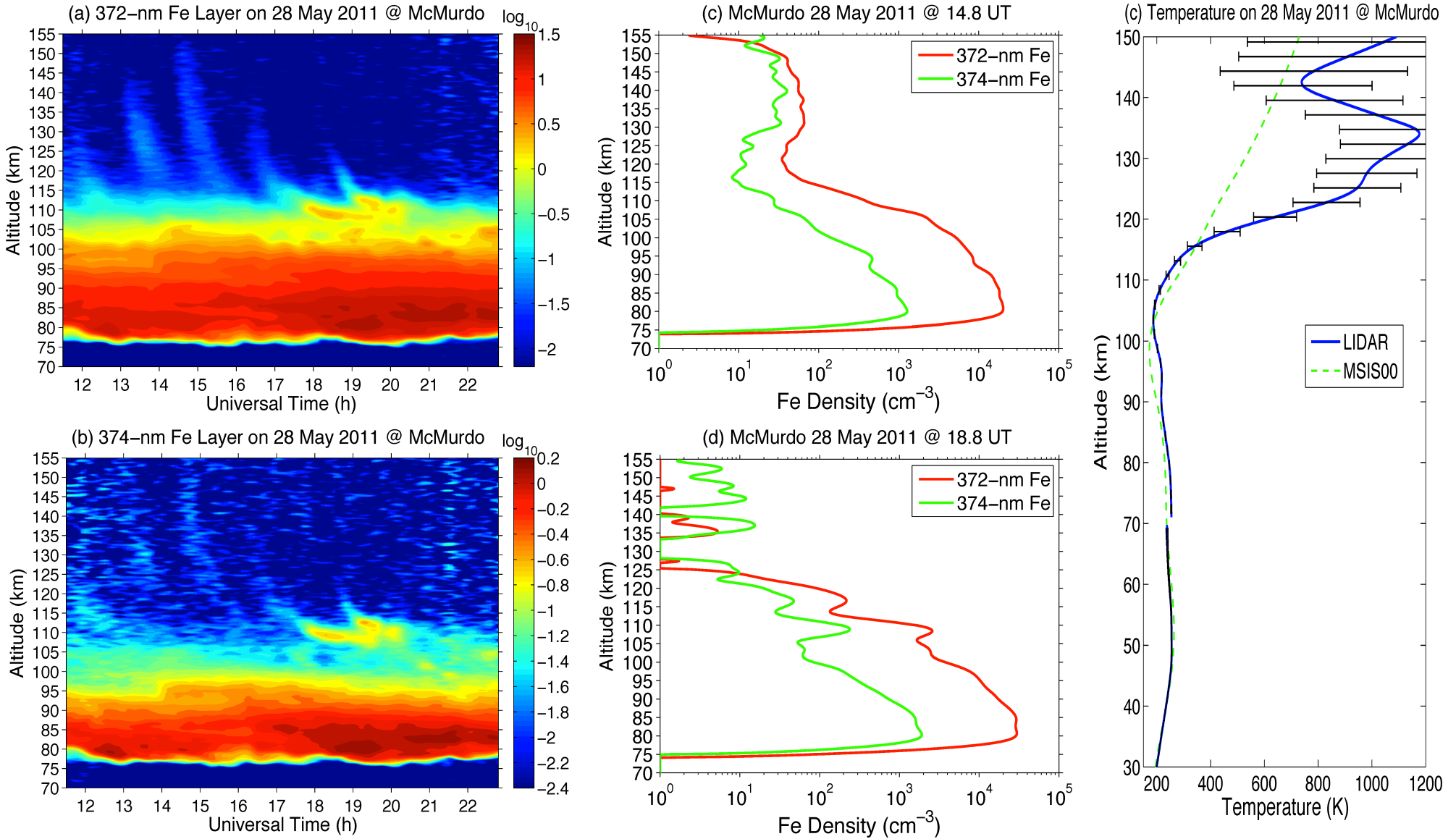
$$= hc \times 416 \times 100(J)$$

$$\Delta E / k_B = 598.43K$$

For $T = 200$,

$$\frac{N(J=4)}{N(J=3)} = \frac{9}{7} e^{598.43/200} = 25.6$$

Example Lidar Signals from Antarctica



Taken from [Chu et al., Geophysical Research Letters, 2011]



Backscatter Cross-Section Comparison

Physical Process	Backscatter Cross-Section	Mechanism
Mie (Aerosol) Scattering	$10^{-8} - 10^{-10} \text{ cm}^2\text{sr}^{-1}$	Two-photon process Elastic scattering, instantaneous
Atomic Absorption and Resonance Fluorescence	$10^{-13} \text{ cm}^2\text{sr}^{-1}$	Two single-photon process (absorption and spontaneous emission) Delayed (radiative lifetime)
Molecular Absorption	$10^{-19} \text{ cm}^2\text{sr}^{-1}$	Single-photon process
Fluorescence from molecule, liquid, solid	$10^{-19} \text{ cm}^2\text{sr}^{-1}$	Two single-photon process Inelastic scattering, delayed (lifetime)
Rayleigh Scattering (Wavelength Dependent)	$10^{-27} \text{ cm}^2\text{sr}^{-1}$	Two-photon process Elastic scattering, instantaneous
Raman Scattering (Wavelength Dependent)	$10^{-30} \text{ cm}^2\text{sr}^{-1}$	Two-photon process Inelastic scattering, instantaneous



Summary (1)

- ❑ Numerous physical processes are involved in lidars, including the interaction between light and objects, and the light transmission through the atmosphere or other medium.
- ❑ Main physical processes for interaction between light and objects include elastic and inelastic scattering, absorption and differential absorption, resonance fluorescence, laser induced fluorescence, Doppler effect, Boltzmann distribution, reflection from target or surface, and multiple scattering. There are large differences in scattering cross sections for various physical processes involved in lidar.
- ❑ Fundamentals to understand atomic structures and energy levels are related to the interactions inside and outside an atom, including electrostatic interaction, electron spin, spin-orbit angular momentum coupling, nuclear spin, external field, etc. Molecular structures and energy levels further include interactions among atoms within a molecule.



Summary (2)

- ❑ Interactions between light and objects are the basis of lidar remote sensing, because it is these interactions that modify light properties so that the light can carry away the information of the objects.
- ❑ Understanding these physical processes precisely is the key to successful lidar simulations and applications.
- ❑ Lidar equation may change its form to best fit for each particular physical process and lidar application.

Any questions on the atomic and molecular spectroscopy?

Our Textbook – Chapter 3 for elastic scattering and polarization
Chapter 4 for differential absorption
Chapters 5 & 7 for resonance fluorescence, Boltzmann, Doppler
Chapter 6 for laser-induced fluorescence
Laser monitoring of the atmosphere (Hinkley) Ch. 3 and 4
Laser Remote Sensing (Measures) Chapter 4
Lidar (Ed. Weitkamp) Chapters 2 and 3