

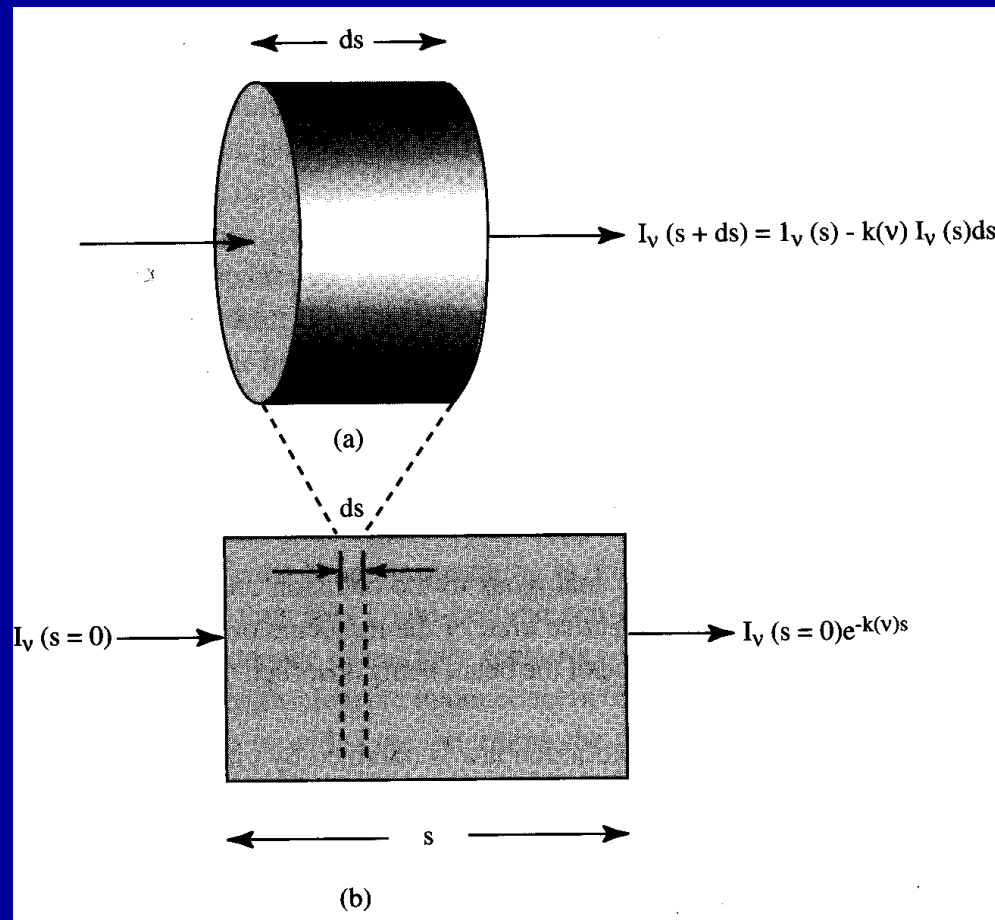
Lesson 4

Radiation Extinction & Scattering

Basic radiative processes

- There are three radiation-matter interactions, absorption, emission and scattering
- We can consider the radiation field in two ways, classical and quantum.
- Classical – the electromagnetic field is a continuous function of space and time, with a well defined electric and magnetic field at every location and instant of time
 - More for scattering
- Quantum – the radiation field is a concentration of discrete values of energy, $h\nu$.
 - More for absorption

The extinction law



Consider a small element of an absorbing medium, ds , within the total medium s .

Extinction Law

- The extinction law can be written as

$$dI_{\nu} = -k(\nu) I_{\nu} ds$$

- The constant of proportionality is defined as the extinction coefficient. k can be defined in three ways.

(1) by the length of the absorbing path with the gas at one atmosphere pressure

$$k(\nu) = \frac{dI_{\nu}}{I_{\nu} ds} \quad (m^{-1})$$

Extinction coefficient

- By mass

$$k_m(\nu) = -\frac{dI_\nu}{I_\nu ds} = -\frac{dI_\nu}{I_\nu dM} \quad [m^2 \cdot Kg^{-1}]$$
$$dM = \rho ds$$

- or by concentration

$$k_n(\nu) = -\frac{dI_\nu}{I_\nu nds} = -\frac{dI_\nu}{I_\nu dN} \quad (m^2)$$
$$dN = nds$$

Optical depth

- Normally we are interested in the total extinction over a finite distance (path length)

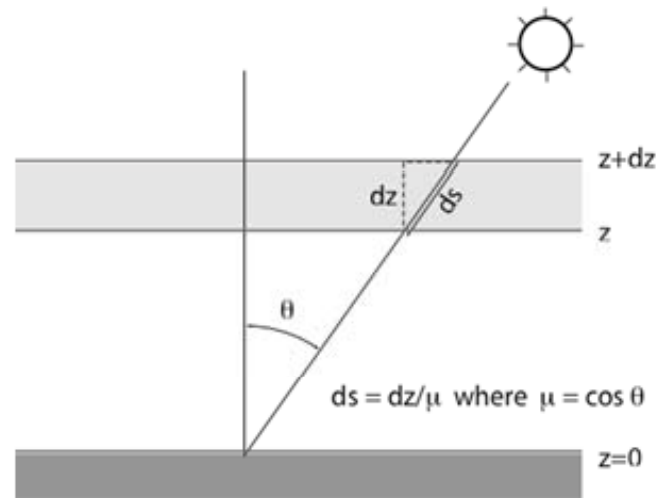
$$\tau_s(\nu) = \int_0^s ds' k(\nu) = \int_0^s ds' k_m(\nu) \rho = \int_0^s ds' k_n(\nu) n$$

Where $\tau_s(\nu)$ is the extinction optical depth

- The integrated form of the extinction equation becomes

$$I_\nu(s, \bar{\Omega}) = I_\nu(0, \bar{\Omega}) \exp[-\tau_s(\nu)]$$

Oblique path



$$dI = -I \cdot k \cdot ds = -I \cdot k \cdot \frac{dz}{u}$$

Optical thickness: $\tau = k \cdot z$
(often used definition)

$$u \frac{dI}{d\tau} = -I$$

Integral form of radiative transfer equation: Beer's law

Goal is to get $I(\tau)$

Coordinate system

Along path

Vertical

Equation to solve:

$$\frac{dI}{d\tau} = -I$$

$$u \frac{dI}{d\tau} = -I$$

Initial condition:

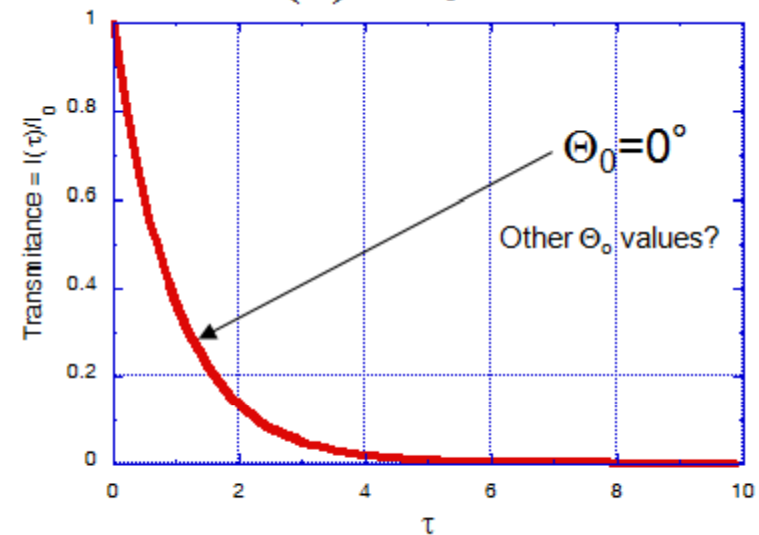
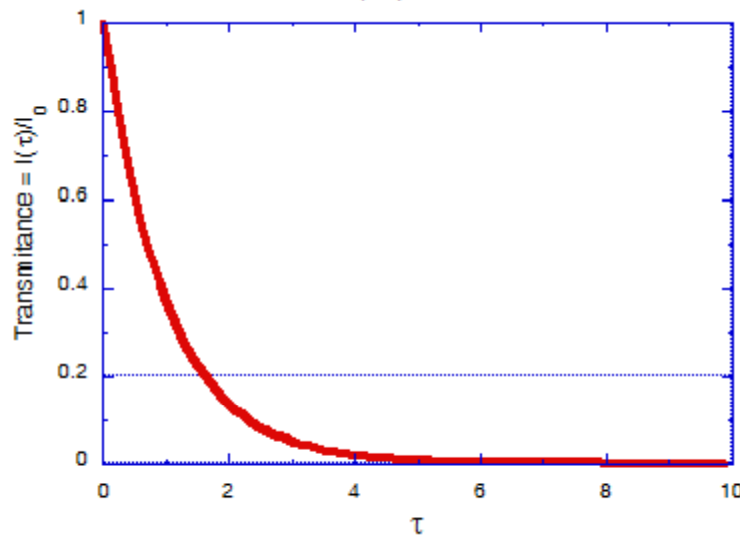
$$I(\tau = 0) = I_0$$

$$I(\tau = 0) = I_0$$

Solution:

$$I(\tau) = I_0 \cdot e^{-\tau}$$

$$I(\tau) = I_0 \cdot e^{-\frac{\tau}{u}}$$



Extinction = scattering + absorption

- Extinction really consists of two distinct processes, scattering and absorption, hence

$$\tau_s(\nu) = \tau_{sc}(\nu) + \tau_a(\nu)$$

where

$$\tau_{sc}(\nu) = \sum_i \int_0^s ds^i \sigma^i(\nu, s')$$

$$\tau_a(\nu) = \sum_i \int_0^s ds^i \alpha^i(\nu, s')$$

Scattering

Air molecules scatter light as dipoles

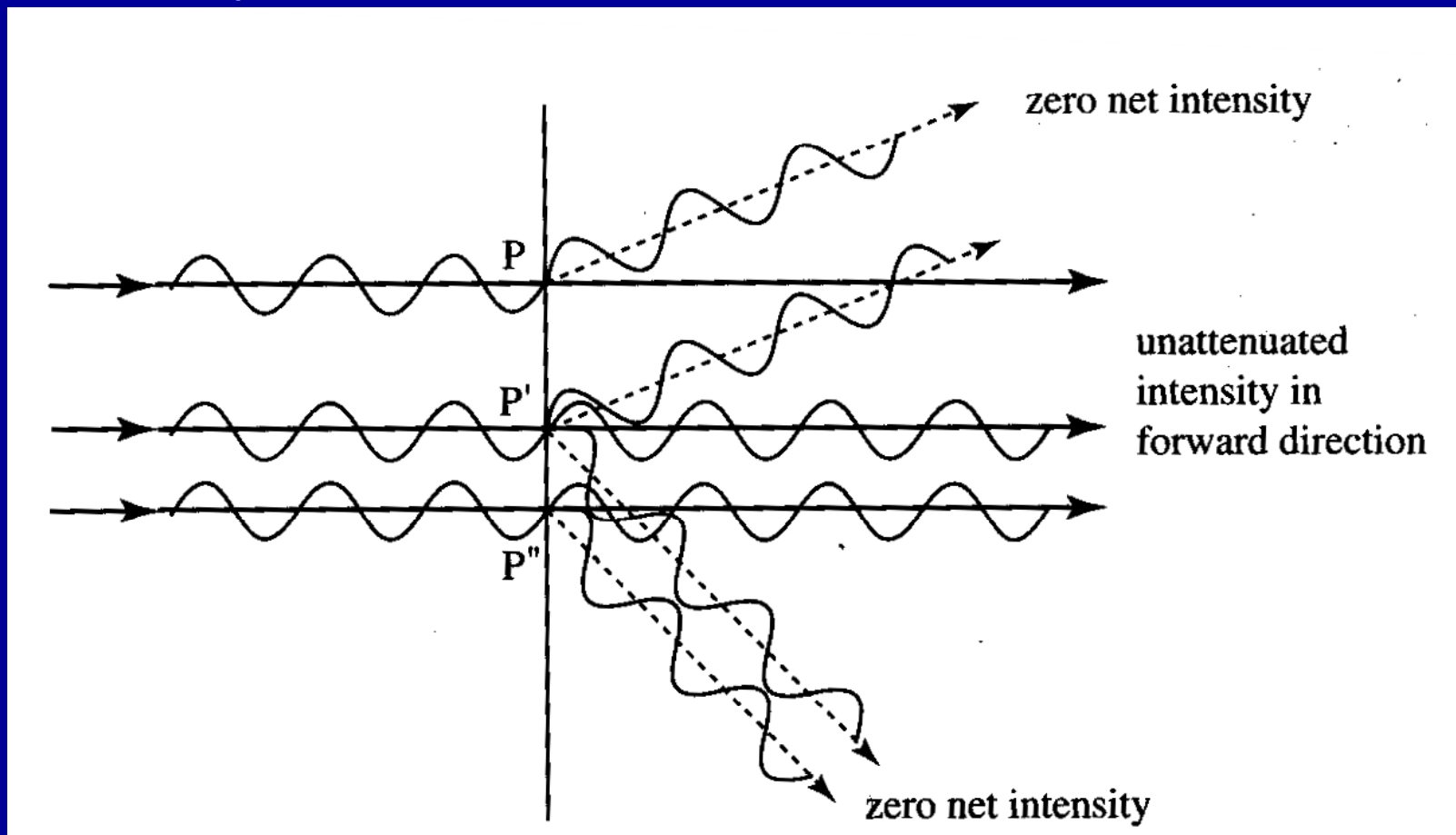


Dipole induced

We use dipole and molecule nearly synonymously → molecules can be approximated as dipoles. If sufficiently small, any particle can be approximated by a dipole oscillator

Scattering of radiation fields

- Radiation fields scattered from the points P'' and P' are 90 degrees different in phase and therefore interfere destructively.



Terminology of Scattering

- Conservative scattering
 - Scattering without absorption (e.g. Rayleigh)
- Nonconservative scattering:
 - Scattering with absorption
- Coherent scattering
 - No/little change to the frequency
- Inelastic scattering (Raman lidar)
 - Scattering with an exchange of internal energy of the medium with that of radiation

Lorentz theory of radiation-matter interactions

- Neutral atoms consist of electrons (negative charges) and nucleus (positive charge).
- Bound together by elastic forces – Hooke's Law.
- Combined with the Maxwell theory of the electromagnetic field
- Classical theory

Scattering from Damped Simple Harmonic Oscillator

- Assume that a molecule is a simple harmonic oscillator with a single harmonic oscillation frequency ω_0 ($2\pi\nu$)
- When irradiated by monochromatic electromagnetic wave of frequency ω_0 , the electron undergoes an acceleration, while the nucleus, being massive, is assumed not to move.
- An accelerating charge gives rise to electromagnetic radiation.

Damped Harmonic Oscillator

- Without energy loss the oscillator would keep its motion indefinitely – forward beam would be unchanged. In reality we see absorption – an energy loss.
- Can only occur if there is some damping force acting on the oscillator. The classical damping force is given by:

$$F = -m_e \gamma v \quad \text{where} \quad \gamma = \frac{e^2 \omega_0^2}{6\pi \epsilon_0 m_e c^3}$$

e is the electronic charge

ϵ_0 is the vacuum permittivity

m_e is the mass of the electron.

Oscillator Strength

- In the classical theory the integrated cross section is a constant. Under the quantum theory there is usually more than one resonant frequency, and each resonance has an integrated cross section given by the above term, but multiplied by a constant f .
- f is called the oscillator strength

$$O_n^{rea}(\nu) = \frac{e^2 f_i}{4m_e \epsilon_0 c} \Phi_L(\nu) \equiv S_i \Phi_L(\nu)$$

Where S_i is called the line strength;

Lorentz profile

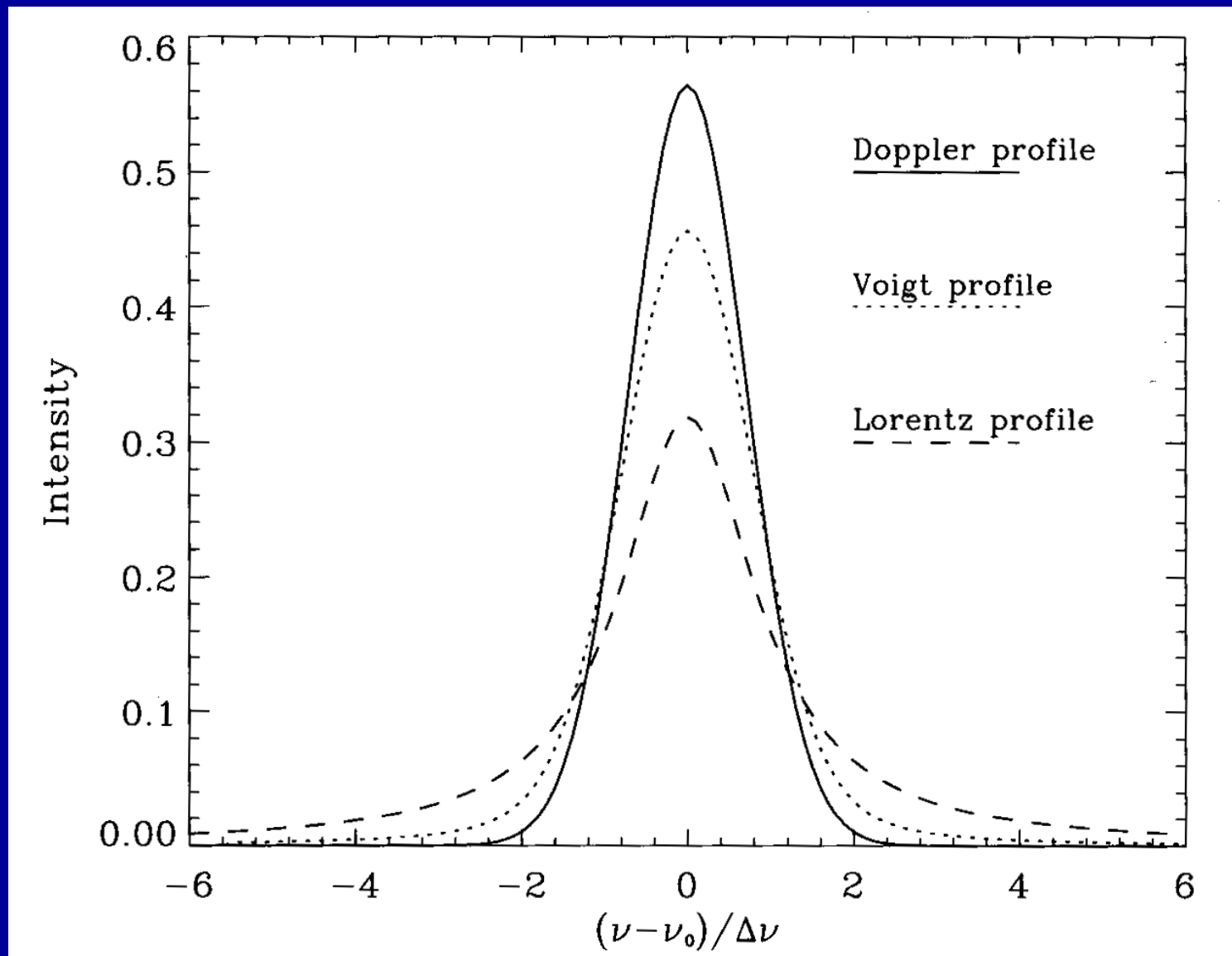
- The frequency dependent part of the equation is called the Lorentz profile

$$\Phi_L(\nu) = \frac{\gamma / 4\pi}{\pi \left[(\nu_0 - \nu)^2 + (\gamma / 4\pi)^2 \right]}$$

Since the Lorentz profile is normalized we find by integrating over all frequencies

$$\int_0^{\infty} d\nu \sigma_n^{res}(\nu) = \frac{e^2}{4m_e \epsilon_0 c}$$

Comparison of line shapes



Scattering is governed by
refractive index and
particle size distribution

Complex Refractive Indices of Liquid Water

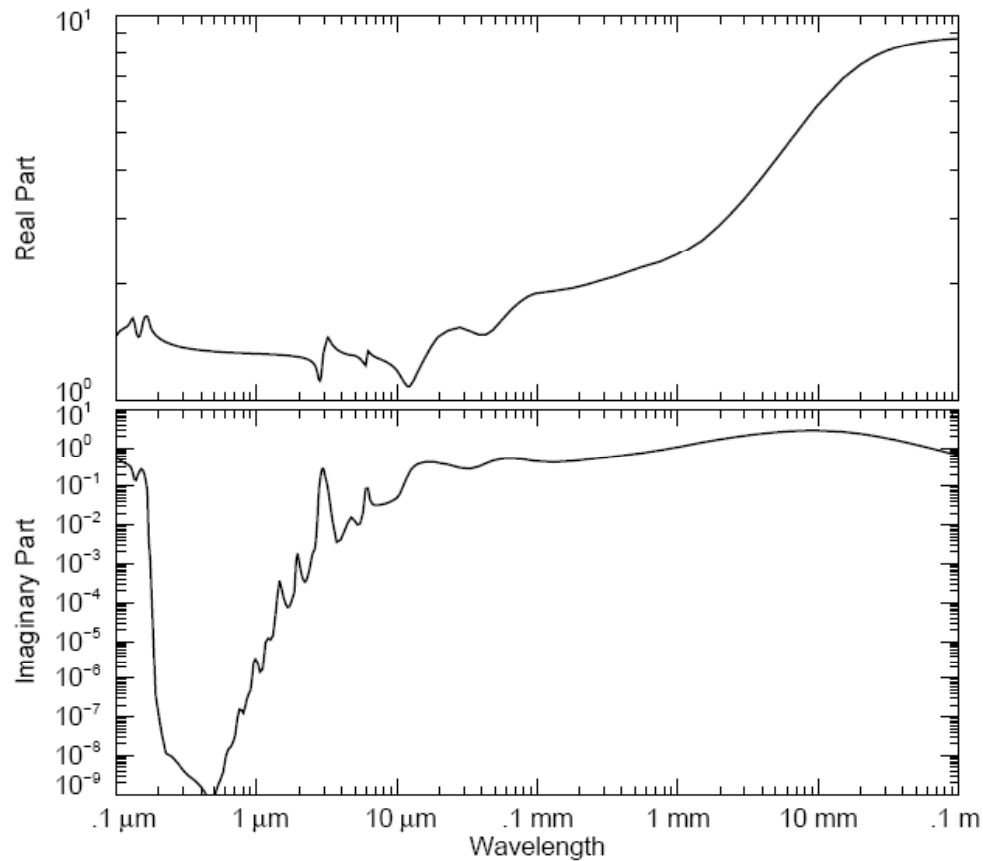


Figure 3.8: Optical constants (complex refractive index) of liquid water taken from the compilation by Querry et al. (1991) cited at the end of Chapter 2.

Size Distribution Matters to Scattering: Lognormal Function

Theoretical size distributions

Gamma and lognormal distributions are used to represent particle size distributions. Aerosol size distributions often represented by a sum of three lognormal distributions (which characterize the three production modes).

$$\text{log normal: } n(r) = \frac{N}{\sigma\sqrt{2\pi}} \frac{1}{r} \exp\left[-\frac{(\ln(r/r_0))^2}{2\sigma^2}\right]$$

r_0 is the modal radius (in $\ln r$), σ is the standard deviation of $\ln r$.

$$\text{Moments: } \int_0^{\infty} r^k n(r) dr = N r_0^k \exp\left(k^2 \sigma^2 / 2\right)$$

Problem

For large size parameters, say $x > 50$, the extinction efficiency asymptotes to $Q_{ext} = 2$. Relate the volume extinction coefficient, β_{ext} , of a particle size distribution to the liquid water content (LWC) and the effective radius.

Size Distribution Matters to Scattering: Gamma Function

$$n(r) = \frac{Nb^{\alpha+1}}{\Gamma(\alpha+1)} r^\alpha \exp(-br) \text{ where } r_c = \alpha/b \text{ is the modal radius and } \alpha \text{ controls the width.}$$

$$\text{Moments: } \int_0^\infty r^k n(r) dr = Nb^{-k} \frac{\Gamma(\alpha+k+1)}{\Gamma(\alpha+1)}$$

$$\text{Effective radius: } r_{eff} = (\alpha+3)/b$$

$$\text{Effective variance: } v_{eff} = 1/(\alpha+3)$$

$$\text{Liquid water content: } LWC = \frac{4\pi}{3} \rho N b^{-3} (\alpha+3)(\alpha+2)(\alpha+1)$$

The cloud drop effective radius

weighted mean of the size distribution of cloud droplets.^[1]

The term was defined in 1974 by [James E. Hansen](#) and [Larry Travis](#) as the ratio of the third to the second moment of a droplet size distribution to aid in the inversion of remotely sensed data.^[2]

Physically, it is an area weighted radius of the cparticles.

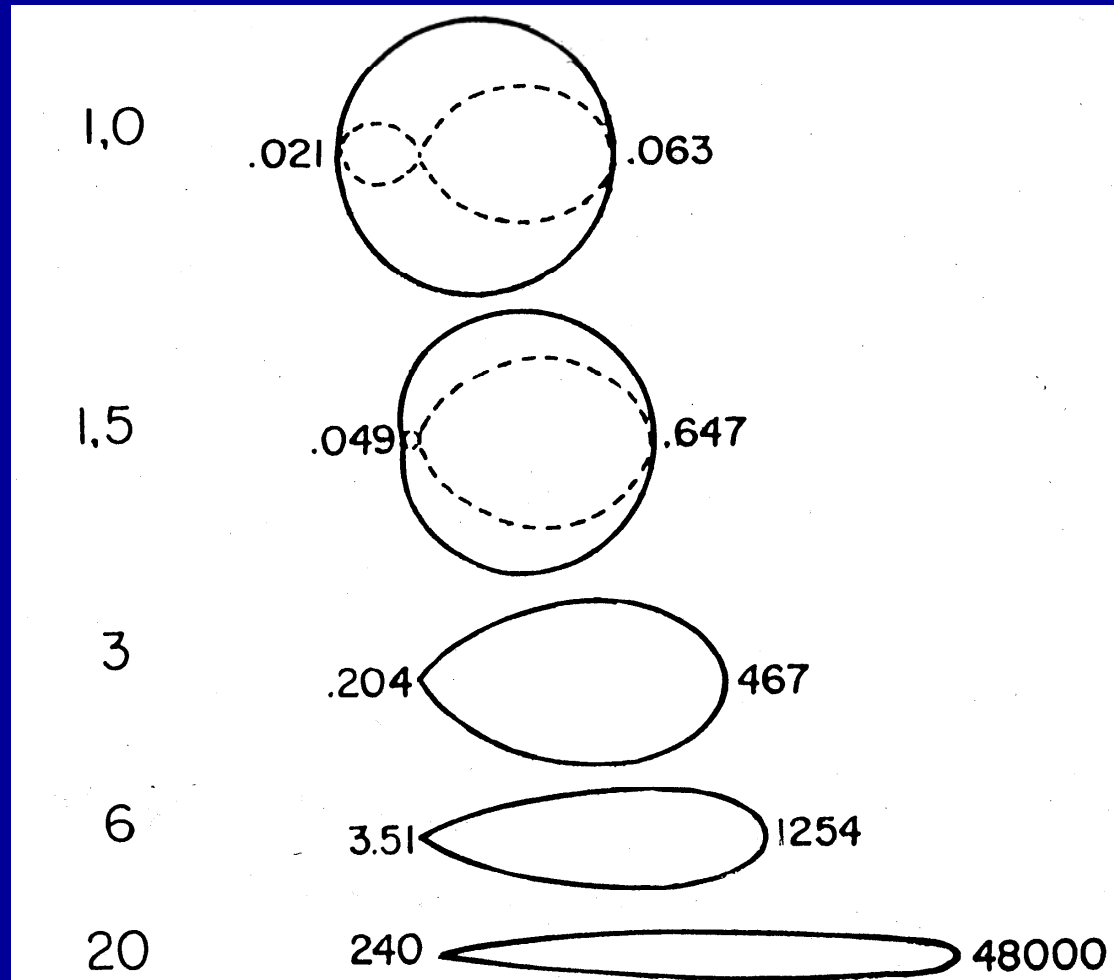
Mathematically, this can be expressed as ■

$$r_e = \frac{\int_{r_1}^{r_2} \pi \cdot r^3 \cdot n(r) dr}{\int_{r_1}^{r_2} \pi \cdot r^2 \cdot n(r) dr}$$

Scattering phase function

- So far we have ignored the directional dependence of the scattered radiation - phase function
- Let the direction of incidence be Ω' , and direction of observation be Ω . The angle between these directions is $\cos\Theta = \Omega' \cdot \Omega$. Θ is the scattering angle.
- If Θ is $< \pi/2$ - forward scattering
- If Θ is $> \pi/2$ - backward scattering

Phase diagrams for aerosols



Phase diagrams for different values of the ratio of the aerosol radius to the wavelength of the incident radiation (left hand column)

Scattering phase function

- In polar coordinates

$$\cos\Theta = \cos\theta' \cos\theta + \sin\theta' \sin\theta \cos(\phi' - \phi)$$

- We define the phase function as follows

$$p(\cos\Theta) = \frac{n\sigma_n(\cos\Theta)}{n \int_{4\pi} d\omega \sigma_n(\cos\Theta)} \quad (sr^{-1})$$

The normalisation is

$$\int_{4\pi} d\omega \frac{p(\cos\Theta)}{4\pi} = \int_0^{2\pi} d\phi \int_0^\pi d\theta \sin\theta \frac{p(\theta', \phi'; \theta, \phi)}{4\pi} = 1$$

Asymmetry Factor

Definition

It is a measure of the preferred scattering direction (forward or backward) .
In radiative transfer studies, asymmetry factor 'g' is equal to the mean value of μ (the cosine of the scattering angle), weighted by the angular scattering phase function $P(\mu)$.
Phase function is defined as the energy scattered per unit solid angle in a given direction to the average energy in all directions.

$$g_{\lambda} = \frac{1}{2} \int_{-1}^{+1} \mu P_{\lambda}(\mu) d\mu$$

The asymmetry factor approaches

+1 for scattering strongly peaked in the forward direction and
-1 for scattering strongly peaked in the backward direction.

0 indicates scattering directions evenly distributed

i.e isotropic scattering (e.g scattering from small particle

$g < 0$ scattering in the backward direction

(i.e scattering angle > 90 deg., often backscattering is referred to scattering at 180 deg.

$g > 0$ scattering in the forward direction

(i.e scattering angle < 90 , often forward-scattering is referred to scattering at 0 deg.

For larger size or Mie particles, g is close to +1.

Single Scattering Albedo and Refractive Index

The ratio of scattering and total extinction coefficient:

$$\omega = \frac{S}{\alpha + S}$$

Refractive index: $m = m_r + i m_i$

Imaginary part (m_i): $dI = -I d\tau_a$
If only absorption is considered: $\tau_a = \int \alpha \cdot \rho \cdot ds = \int k \cdot ds$

(α = absorption coefficient; ρ = density; s = path, k = absorption coefficient)

$$\alpha = \frac{2\pi \cdot m_i}{\lambda}$$

The value of m_i depends on how easy it is to bounce electrons to higher energy levels so that they don't fall back.

Usually: small values + sharp peaks at a few wavelengths
(though learned about widening of absorption spectra)

Single scattering albedo

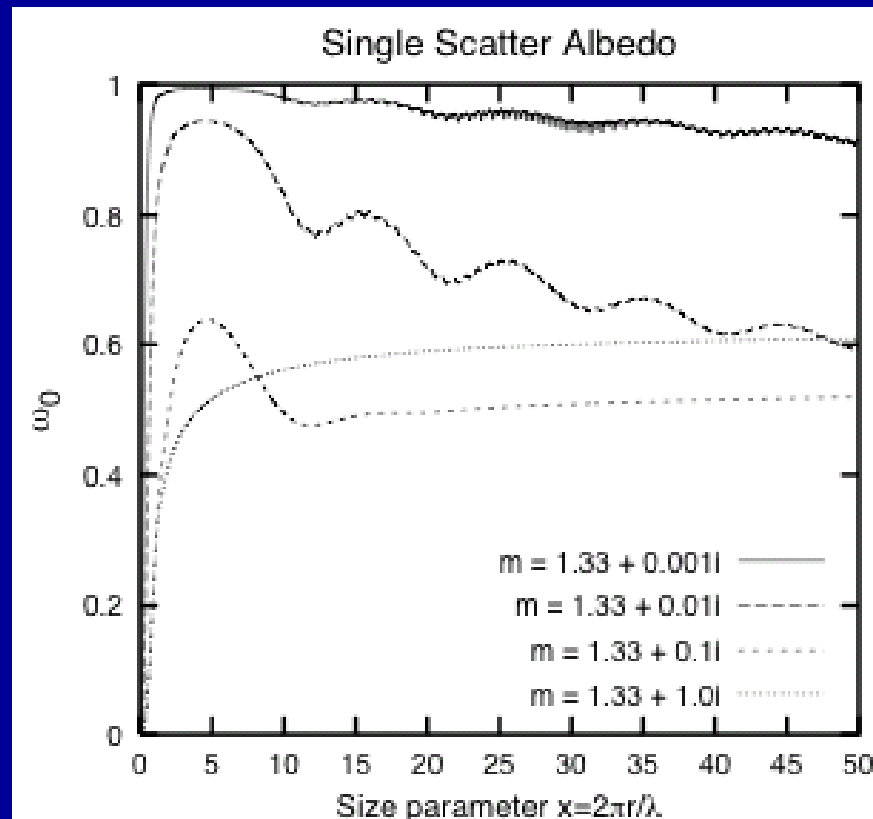
Upper bound: 1.0

Lower bound for large particles: 0.5

Typical values for droplets at visible wavelengths: just below 1.0

Some aerosols contain mix of water and carbon -> lower values

Wavelength-dependence: decrease with size



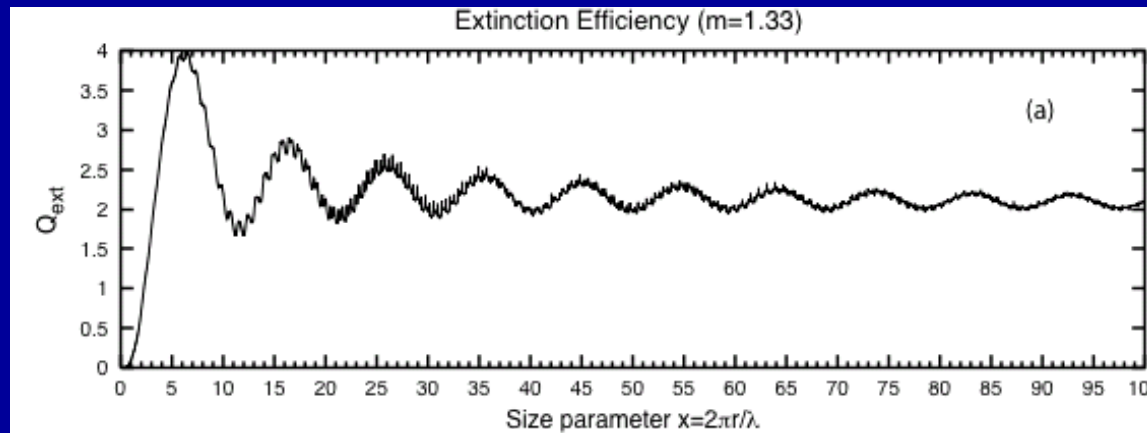
Scattering by clear air 2

We know that

$$\sigma = \int_0^{\infty} Q(r, \lambda) \cdot n(r) \cdot \pi r^2 \cdot dr = \int_0^{\infty} Q(x) \cdot n(r) \cdot \pi r^2 \cdot dr$$

$$x = \frac{2\pi r}{\lambda} \quad \text{Size Parameter}$$

In clean air, r remains constant, but λ of interest may vary



Which part of $Q(x)$ curve applies?

$$Q(x) \propto x^4 \propto \frac{1}{\lambda^4}$$

$\tau \approx 0.2$ in blue

$\tau \approx 0.03$ in red

Easy-to-calculate formulas available (will have code)

Rayleigh Scattering by Air Molecules

Lord Rayleigh
John William Strutt
(third Baron Rayleigh)
1842-1919
Essex, Cambridge

Nobel Prize in Physics in 1904
"for his investigations of the densities
of the most important gases
and for his discovery of argon
in connection with these studies"



Rayleigh scattering

- If the size of scatter is much less than the wavelength of light, the scattering cross section for a damped simple oscillator becomes

$$\sigma_n^{RAY} = \frac{e^4 \omega^4}{6\pi m_e^4 \epsilon_0^2 c^4 \omega_0^4} = \frac{1}{6\pi} \left(\frac{\omega}{c} \right)^4 \left(\frac{e^2}{m_e \epsilon_0 \omega_0^2} \right)$$

- The molecular polarizability is defined as

$$\alpha_p = \frac{e^2}{4\pi m_e \epsilon_0 \omega_0^2} \text{ for } \omega \ll \omega_0$$

Rayleigh scattering

- Transforming from angular frequency to wavelength we get

$$\sigma_n^{RAY}(\lambda) = \frac{8\pi}{3} \left(\frac{2\pi}{\lambda} \right)^4 \alpha_p^2$$

Rayleigh scattering

- The polarizability can be expressed in terms of the real refractive index, m_r

$$\alpha_p = (m_r - 1) / 2\pi n$$

$$\sigma^{RAY}(\lambda) = \sigma_n^{RAY} n = 32\pi^3 (m_r - 1)^2 (m^{-1})$$

where $\sigma^{RAY}(\lambda)$ is the scattering coefficient (per atmosphere)

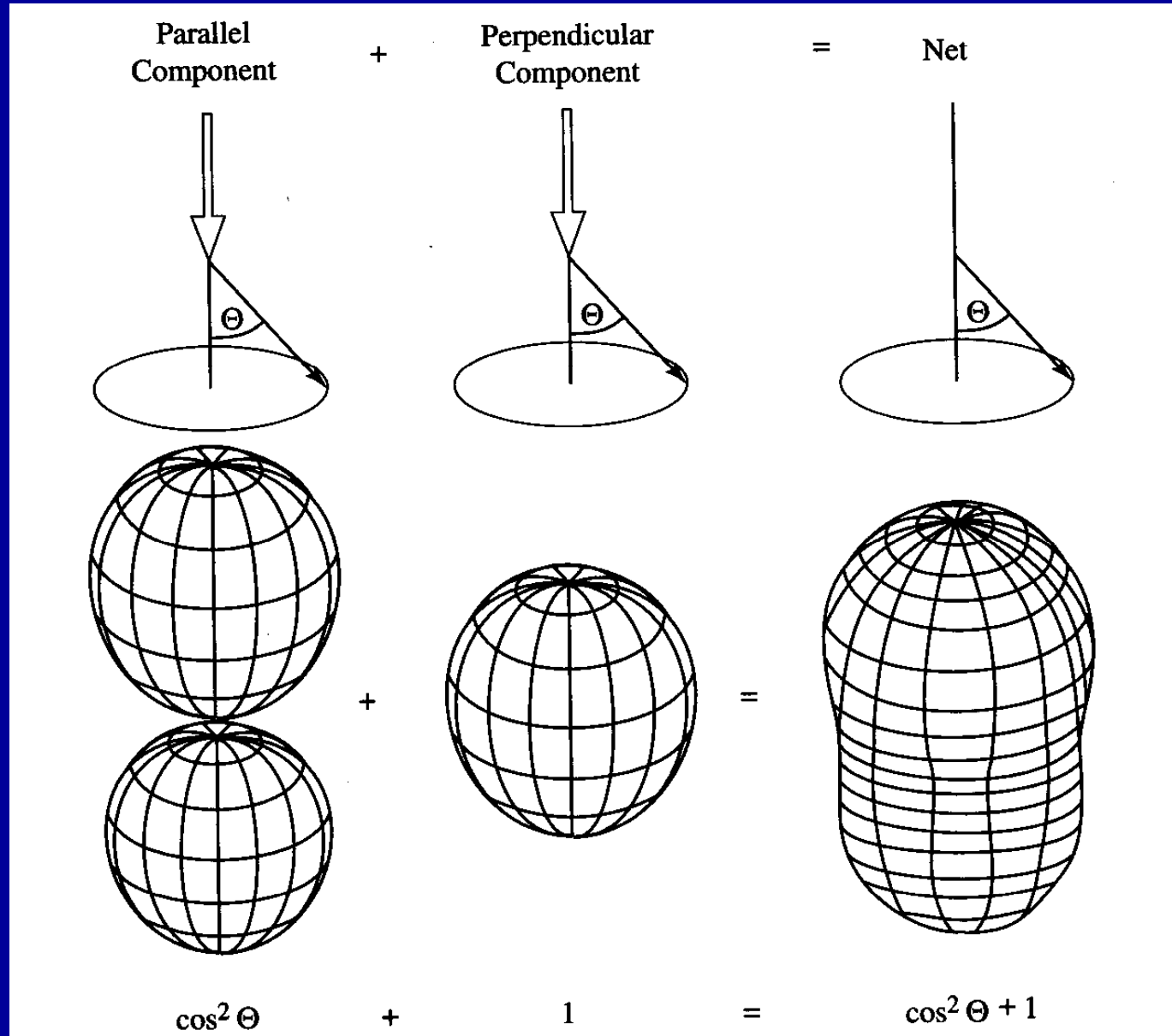
- m_r varies with wavelength, so the actual cross section deviates somewhat from the λ^{-4} dependence

Rayleigh scattering phase function

$$\frac{1}{4\pi} \int_{4\pi} d\omega (1 + \cos^2 \theta) = \frac{1}{4\pi} \int_0^{2\pi} d\phi \int_0^{2\pi} d\theta \sin \theta (1 + \cos^2 \theta) = \frac{4}{3}$$

$$P_{ray}(\Theta) = \frac{3}{4} (1 + \cos^2 \Theta)$$

Phase diagram for Rayleigh scattering



Rayleigh scattering

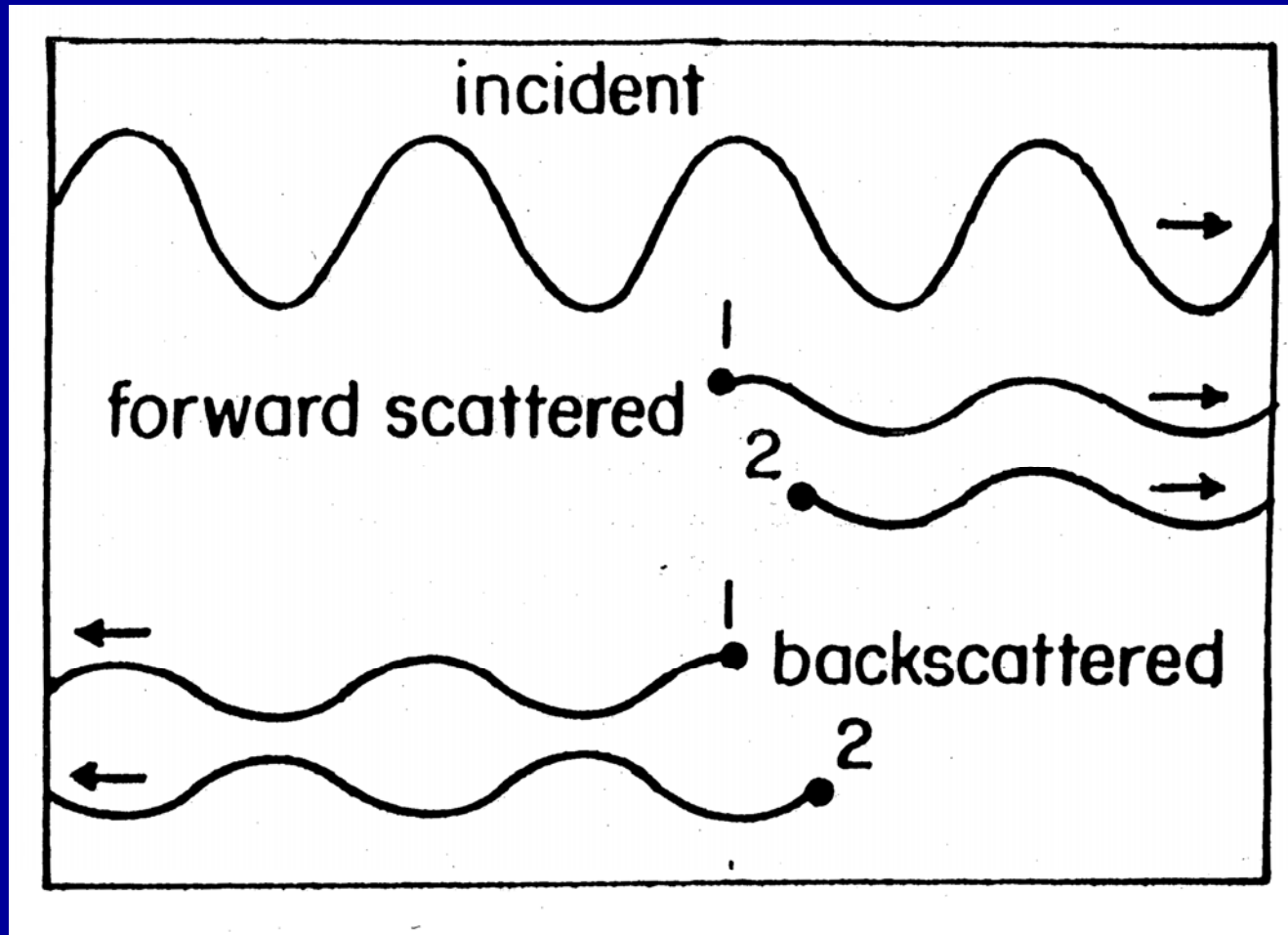
λ , nm	σ , cm ²	τ , surface	Exp(- τ)
300	6.00 E-26	1.2	0.301
400	1.90 E-26	0.38	0.684
600	3.80 E-27	0.075	0.928
1000	4.90 E-28	0.0097	0.990
10,000	4.85 E-32	9.70 E-7	0.999

- Sky appears blue at noon, red at sunrise and sunset - why?

Lorenz-Mie-Debye Theory

- Mie theory, also called Lorenz-Mie theory or Lorenz-Mie-Debye theory, is an analytical solution of Maxwell's equations for the scattering of electromagnetic radiation by spherical particles (also called Mie scattering) in terms of infinite series. The Mie solution is named after its developer, German physicist Gustav Mie. However, others like Danish physicist Lorenz preceded him independently developed the theory of electromagnetic plane wave scattering by a dielectric sphere. The term "Mie solution" is sometimes used more generically for any analytical solution in terms of infinite series,

Schematic of scattering from a large particle



In the diagram above 1 and 2 are points within the particle. In the forward direction the induced radiation from 1 and 2 are in phase. However in the backward direction the two induced waves can be completely out of phase.

Mie Theory

Scattering by Spherical Particles

- Mie theory: scattering by arbitrary homogeneous sphere illuminated by a plane wave
- Mie scattering is a theory (one of many), not a physical process
- Scattering by a sphere can also be determined by Fraunhofer theory, geometrical optics, anomalous diffraction, coupled-dipole, ... (among many others)
- No distinct boundary between Mie and Rayleigh scatterers; Mie theory includes Rayleigh theory (applicable as $x \rightarrow 0$)
- Mie scattering by cylinders, spheroids, coated spheres? Mie never considered it; not “Mie theory.”

Mie-Debye Scattering by Particles

- Unlike absorption, scattering can be apportioned into directions:
- *differential scattering cross section is the contribution to the total scattering cross section C_{sca} from scattering into a unit solid angle in each direction:*

$$C_{sca} = \int_{4\pi} \frac{dC_{sca}}{d\Omega} d\Omega$$

- ***Scattering coefficient β of a suspension of N identical particles per unit volume:***

$$\beta = NC_{sca}$$

- sum of the absorption and scattering cross sections is called the *extinction cross section: $C_{ext} = C_{abs} + C_{sca}$;*

scattering and extinction are sometimes normalized by their geometrical (projected) cross-sectional areas G to yield *dimensionless efficiencies or efficiency factors for scattering and extinction:*

- $Q_{ext} = Q_{sca} + Q_{abs}$

- Expand the incident, scattered, and internal EM fields in a series of vector spherical harmonics (general solutions to the equations of EM field in spherical harmonics)
- Coefficients of the expansion functions are

$$C_{sca} = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \Re \left\{ |a_n|^2 + |b_n|^2 \right\} \quad C_{ext} = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \Re \{ a_n + b_n \}$$

Scattering coefficients:

$$b_n = \frac{[mD_n(mx) + n/x] \Psi_n(x) - \Psi_{n-1}(x)}{[mD_n(mx) + n/x] \xi_n(x) - \xi_{n-1}(x)} \quad a_n = \frac{[D_n(mx)/m + n/x] \Psi_n(x) - \Psi_{n-1}(x)}{[D_n(mx)/m + n/x] \xi_n(x) - \xi_{n-1}(x)}$$

$\Psi_n(x)$ and $\xi_n(x)$ are *Riccati-Bessel* functions and the logarithmic derivative is:

$$D_n(x) = \frac{d}{dx} \ln \Psi_n(x)$$

- Size parameter x is $kr = 2\pi r/\lambda$ where r is radius of sphere, k is wavenumber of incident radiation, λ ; m is complex refractive index of sphere **relative** to refractive index of surrounding medium.

Project 1 – Mie Scattering Due Feb 17

1. Read the papers that I will send you to help you understand the Mie theory and codes.

2. Compute Mie scattering optical properties for liquid cloud droplets with number concentration of 100 cm^{-3} , effective radius of $10 \text{ }\mu\text{m}$, and effective variance of 0.1 ($\alpha = 7$ for a gamma distribution) at wavelengths of $0.87 \text{ }\mu\text{m}$, $1.64 \text{ }\mu\text{m}$, $2.13 \text{ }\mu\text{m}$, $11.0 \text{ }\mu\text{m}$. Also calculate the scattering properties for a cloud with $N = 100 \text{ cm}^{-3}$ and $r_{\text{eff}} = 5 \text{ }\mu\text{m}$ at $1.64 \text{ }\mu\text{m}$. Finally, compute the Mie scattering optical properties for a mineral aerosol layer index of refraction $m = 1.56 - 0.01i$ and size distribution $N = 200 \text{ cm}^{-3}$, $r_{\text{eff}} = 0.7 \text{ }\mu\text{m}$, $\alpha = 1$ at $0.65 \text{ }\mu\text{m}$.

3. Plots the phase function, the extinction (km^{-1}), single scattering albedo, asymmetry parameter for the 6 cases.

4. Discuss the results with regard to the variations of the phase function and single scattering albedo with particle size and refractive index, and wavelength.