

Lecture 15.

Light scattering and absorption by atmospheric particulates. Part 2:

Scattering and absorption by spherical particles.

Objectives:

1. Maxwell equations. Wave equation. Dielectrical constants of a medium.
2. Mie-Debye theory.
3. Volume optical properties of an ensemble of particles.

Required Reading:

L02: 5.2, 3.3.2

Additional/Advanced Reading:

Bohren, G.F., and D.R. Huffman, Absorption and scattering of light by small particles. John Wiley&Sons, 1983 (Mie theory derivation is given on pp.82-114, a hardcopy will be provided in class)

1. Maxwell equations. Wave equation. Dielectrical constants of a medium.

Maxwell equations connect the five basic quantities the electric vector, \vec{E} , magnetic vector, \vec{H} , magnetic induction, \vec{B} , electric displacement, \vec{D} , and electric current density, \vec{j} : (in cgs system)

$$\begin{aligned}\nabla \times \vec{H} &= \frac{1}{c} \frac{\partial \vec{D}}{\partial t} + \frac{4\pi}{c} \vec{j} \\ \nabla \times \vec{E} &= -\frac{1}{c} \frac{\partial \vec{B}}{\partial t} \\ \nabla \cdot \vec{D} &= 4\pi\rho \\ \nabla \cdot \vec{B} &= 0\end{aligned}\tag{15.1}$$

where c is a constant (wave velocity); and ρ is the electric charge density.

To allow a unique determination of the electromagnetic field vectors, the Maxwell equations must be supplemented by relations which describe the behavior of substances under the influence of electromagnetic field. They are

$$\vec{j} = \sigma \vec{E} \qquad \vec{D} = \epsilon \vec{E} \qquad \vec{B} = \mu \vec{H} \tag{15.2}$$

where σ is called the **specific conductivity**; ϵ is called the **dielectrical constant** (or the **permittivity**), and μ is called the **magnetic permeability**.

Depending on the value of σ , the substances are divided into:

conductors: $\sigma \neq 0$ (i.e., σ is NOT negligibly small), (for instance, metals)

dielectrics (or insulators): $\sigma = 0$ (i.e., σ is negligibly small), (for instance, air, aerosol and cloud particulates)

Let consider the propagation of EM waves in a medium which is

- (a) uniform, so that ϵ has the same value at all points;
- (b) isotropic, so that ϵ is independent of the direction of propagation;
- (c) non-conducting (dielectric), so that $\sigma = 0$ and therefore $\mathbf{j} = 0$;
- (d) free from charge, so that $\rho = 0$.

With these assumptions the Maxwell equations reduce to

$$\begin{aligned}\nabla \times \vec{H} &= \frac{\epsilon}{c} \frac{\partial \vec{E}}{\partial t} \\ \nabla \times \vec{E} &= -\frac{\mu}{c} \frac{\partial \vec{H}}{\partial t} \\ \nabla \cdot \vec{E} &= 0 \\ \nabla \cdot \vec{H} &= 0\end{aligned}\tag{15.3}$$

Eliminating E and H in the first two equations in [15.3] and using the vector theorem, we have

$$\begin{aligned}\nabla^2 \vec{E} &= \frac{\epsilon\mu}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} \\ \nabla^2 \vec{H} &= \frac{\epsilon\mu}{c^2} \frac{\partial^2 \vec{H}}{\partial t^2}\end{aligned}\tag{15.4}$$

The above equations are standard equations of wave motion for a wave propagating with a velocity

$$v = \frac{c}{\sqrt{\epsilon\mu}}\tag{15.5}$$

where c is the speed of light in vacuum.

NOTE: for vacuum: $\mu = 1$ and $\epsilon = 1$ in cgs units, but in SI system μ_0 and ϵ_0 are constants such that $c = 1/\sqrt{\epsilon_0\mu_0}$.

- For most substances (including the air) μ is unity. Thus, the electrical properties of a medium are characterized by the dielectrical constant ϵ .

Refractive index (or **optical constants**) of a medium is defined as

$$m = \sqrt{\epsilon} \quad [15.6]$$

assuming that $\mu=1$.

NOTE: Strictly speaking, ϵ in Eq.[15.6] is the relative permittivity of medium (here it is relative to vacuum).

Refractive index:

- ✓ **The refractive index $m=m_r - im_i$** is commonly expressed as a complex number. The nonzero imaginary part m_i of the refractive index is responsible for absorption of the wave as it propagates through the medium; whereas the real part m_r of the refractive index relates to the velocity of propagation of the EM wave.
- ✓ The refractive index is a strong function of the wavelengths. Each substance has a specific spectrum of the refractive index (see figures 4.6-4.7, Lecture 4)
- ✓ Particles of different sizes, shapes and indices of refraction will have different scattering and absorbing properties.
- ✓ Aerosol particles often consist of several chemical species (called the **internal mixture**). There are several approaches (called **mixing rules**) to calculate the **effective refractive index** m_e of the internally mixed particles using the refractive indices of the individual species (see Lecture 4)

➤ Scattering domains:

Rayleigh scattering: $2\pi r/\lambda \ll 1$ and m is arbitrary (applies to scattering by molecules and small aerosol particles);

Rayleigh-Gans scattering: $\frac{2\pi r}{\lambda} |m - 1| \ll 1$ and $|m - 1| \ll 1$ (not useful for atmospheric application);

Mie-Debye scattering: $2\pi r/\lambda$ and m are both arbitrary but for spheres only (applies to scattering by aerosol and cloud particles)

Geometric optics: $2\pi r/\lambda \gg 1$ and m is real (applies to scattering by large cloud droplets).

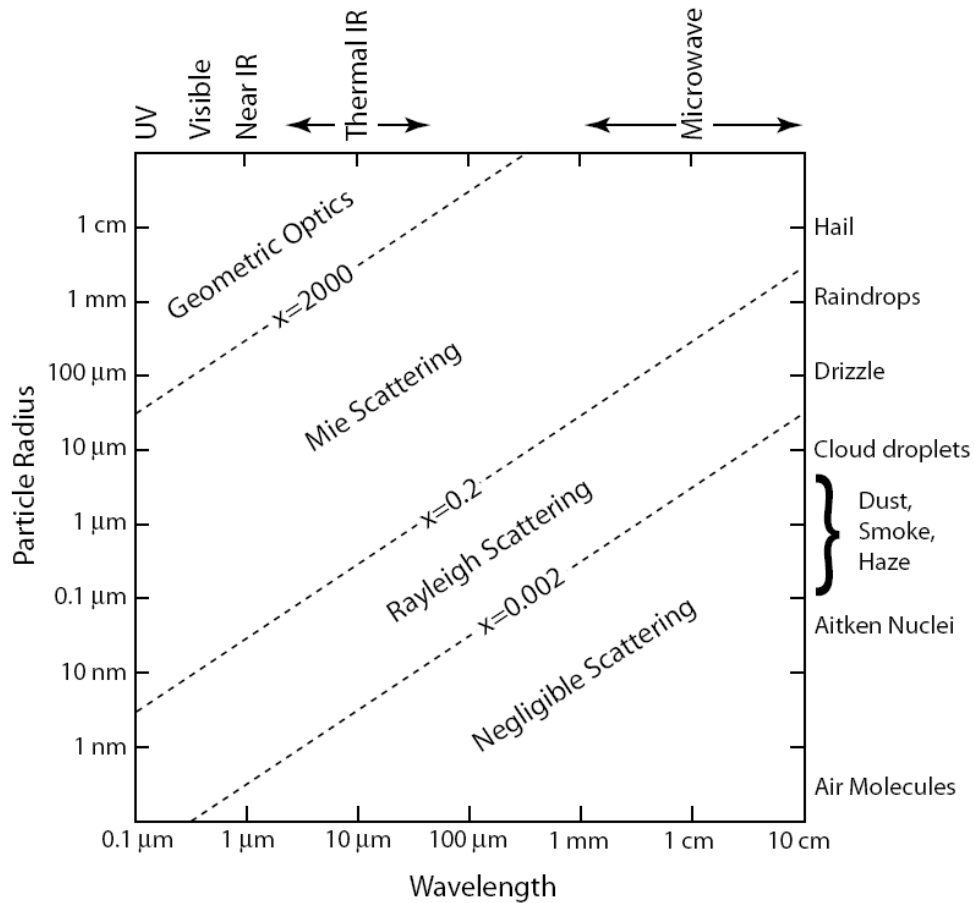


Figure 15.1 Relationship between particle size, radiation wavelength and scattering behavior for atmospheric particles. Diagonal dashed lines represent rough boundaries between scattering regimes.

2. Mie-Debye theory.

NOTE: Mie-Debye theory is often called **Mie theory** or **Lorentz-Mie theory**.

Mie theory outline:

Assumptions:

- i) Particle is a **sphere** of radius r
- ii) Particle is **homogeneous** (therefore it is characterized by a **refractive index** $m=m_r - im_i$ at a given wavelength);

NOTE: Mie theory requires the relative refractive index = refractive index of a particle divided by the refractive index of a medium. But for air m is about 1, so one needs to know the refractive index of the particle (i.e., refractive index of the material of which the particle is composed).

NOTE: If a particle has complex chemical composition, the effective refractive index must be calculated at a given wavelength.

Strategy:

- 1) Seek a solution of a vector wave equation (Eq.[15.4]) for \vec{E} and \vec{H}

$$\nabla^2 \vec{E} = \frac{\epsilon}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2}$$

with the boundary condition that the tangential component of \vec{E} and \vec{H} be continuous across the spherical surface of a particle. Assumption on the spherical surface of a particle allows solving the vector equation analytically.

- 2) Re-write the wave equation in spherical coordinates and express electric field inside and outside sphere in vector spherical harmonic expansions.

NOTE: Mie theory calculates the electromagnetic field at all points in the particle (called internal field) and at all points of the homogeneous medium in which the particle is embedded. For all practical applications in the atmosphere, light scattering observations are carried out in the far-field zone (i.e., at the large distances from a sphere):

- 3) Apply boundary conditions – match transverse fields at sphere surface to obtain scattered spherical wave **Mie coefficients** a_n and b_n which don't depend on the angles

but depend on size parameter $\mathbf{x} = 2\pi r/\lambda$ (r is the radius of the particle) and variable $\mathbf{y} = \mathbf{x} m$ (m is refractive index of the particle).

4) Use series involving a_n and b_n to obtain **extinction and scattering efficiencies** (Q_e and Q_s).

5) Use series in **Mie angular functions** π_n and τ_n to obtain **scattering amplitude functions** $S_1(\Theta)$ and $S_2(\Theta)$, from which the scattering phase function is derived.

NOTE: Full derivation of Mie theory are given in L02, section 5.2 (and Bohren&Huffman 1983, pp.82-114).

Mie scattering amplitudes

(also called scattering functions) derived from Mie theory are (see Eqs.5.2.78 in L02)

$$S_1(\Theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [a_n \pi_n(\cos \Theta) + b_n \tau_n(\cos \Theta)]$$

$$S_2(\Theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [b_n \pi_n(\cos \Theta) + a_n \tau_n(\cos \Theta)] \quad [15.7]$$

where **Mie coefficients** a_n and b_n are (see Eqs.5.2.74 in L02)

$$a_n(x, y) = \frac{\psi'_n(y)\psi_n(x) - m\psi_n(y)\psi'_n(x)}{\psi'_n(y)\xi_n(x) - m\psi_n(y)\xi'_n(x)} \quad b_n(x, y) = \frac{m\psi'_n(y)\psi_n(x) - \psi_n(y)\psi'_n(x)}{m\psi'_n(y)\xi_n(x) - \psi_n(y)\xi'_n(x)} \quad [15.8]$$

here the prime denotes differentiation; $x = 2\pi r/\lambda$ and $y = x m$;

$\psi_n(\rho) = \sqrt{\frac{\pi\rho}{2}} J_{n+1/2}(\rho)$ and $\xi_n(\rho) = \sqrt{\frac{\pi\rho}{2}} H_{n+1/2}^{(2)}(\rho)$ where $J_{n+1/2}(\rho)$ is the half-integral-order

spherical Bessel function and $H_{n+1/2}^{(2)}$ is the half-integral-order Hankel function of the second kind;

and π_n and τ_n are the **Mie angular functions**

$$\pi_n(\cos \Theta) = \frac{1}{\sin(\Theta)} P_n^1(\cos \Theta)$$

$$\tau_n(\cos \Theta) = \frac{d}{d\Theta} P_n^1(\cos \Theta) \quad [15.9]$$

where P_n^1 are the associated Legendre polynomials (see Appendix E).

In the far-field zone (i.e., at the large distances \mathbf{R} from a sphere), Mie theory gives **the solution of the vector wave equation as**

$$\begin{bmatrix} \mathbf{E}_l^s \\ \mathbf{E}_r^s \end{bmatrix} = \frac{\exp(-i\mathbf{kR} + ikz)}{ikR} \begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix} \begin{bmatrix} \mathbf{E}_l^i \\ \mathbf{E}_r^i \end{bmatrix} \quad [15.10]$$

Eq.[15.10] is a fundamental equation of scattered radiation including polarization in the far field.

$\begin{bmatrix} S_2(\Theta) & S_3(\Theta) \\ S_4(\Theta) & S_1(\Theta) \end{bmatrix}$ is the amplitude scattering matrix (unitless)

For spheres: $S_3(\Theta) = S_4(\Theta) = 0$

Thus, **for spheres** Eq.[15.10] reduces to

$$\begin{bmatrix} \mathbf{E}_l^s \\ \mathbf{E}_r^s \end{bmatrix} = \frac{\exp(-i\mathbf{kR} + ikz)}{ikR} \begin{bmatrix} S_2 & 0 \\ 0 & S_1 \end{bmatrix} \begin{bmatrix} \mathbf{E}_l^i \\ \mathbf{E}_r^i \end{bmatrix} \quad [15.11]$$

where $\exp(ikz)$ is the incident plane wave, and $\frac{\exp(-i\mathbf{kR})}{ikR}$ is the outgoing scattered wave.

Fundamental **extinction formula (or optical theorem)** gives the extinction cross section of a particle

$$\sigma_e = \frac{4\pi}{k^2} \text{Re}[S_{1,2}(0^0)] \quad [15.12]$$

But for the forward direction (i.e. $\Theta = 0^0$) from Eq.[15.7], we have

$$S_1(0^0) = S_2(0^0) = \frac{1}{2} \sum_{n=1}^{\infty} (2n+1)(a_n + b_n) \quad [15.13]$$

Thus, extinction cross section is related to scattering in forward direction.

➤ **Efficiencies (or efficiency factors)** for extinction, scattering and absorption are defined as

$$Q_e = \frac{\sigma_e}{\pi r^2} \quad Q_s = \frac{\sigma_s}{\pi r^2} \quad Q_a = \frac{\sigma_a}{\pi r^2} \quad [15.14]$$

where πr^2 is the particle area projected onto the plane perpendicular to the incident beam.

Mie efficiency factors are derived from the Mie scattering amplitude

$$Q_e = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n + 1) \operatorname{Re}[a_n + b_n] \quad [15.15]$$

$$Q_s = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n + 1)[|a_n|^2 + |b_n|^2] \quad [15.16]$$

and the absorption efficiency can be calculated as

$$Q_a = Q_e - Q_s \quad [15.17]$$

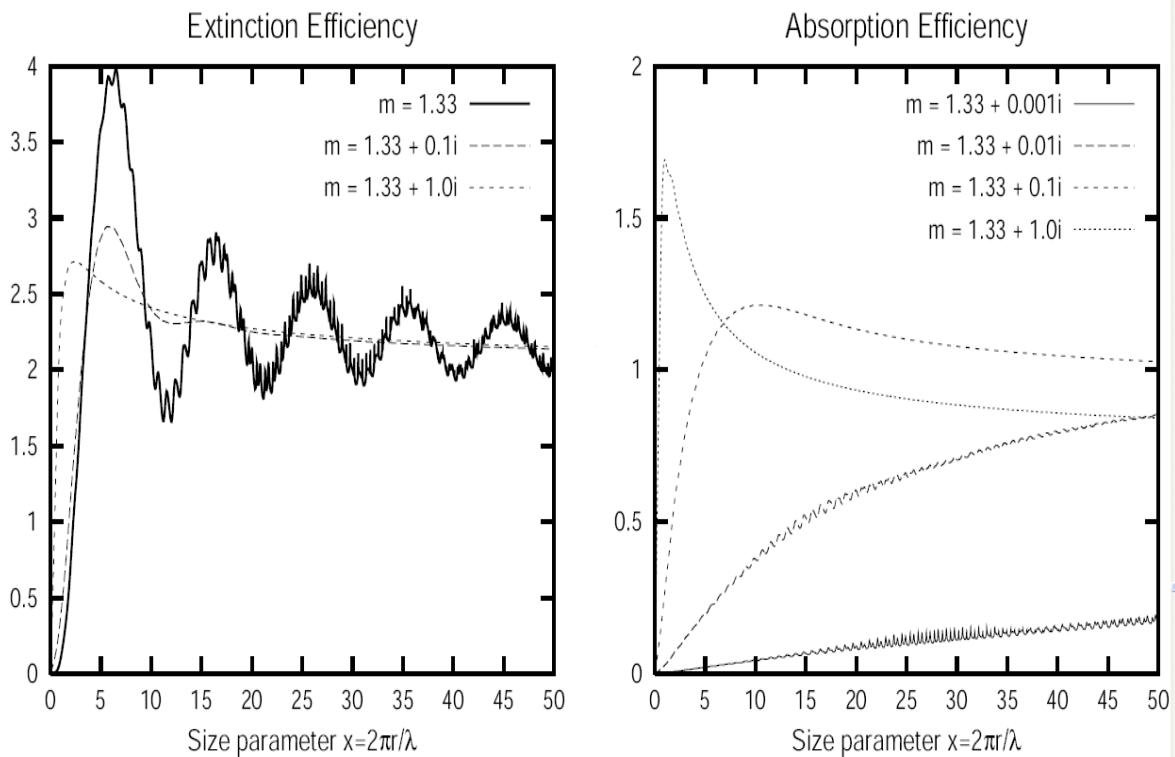


Figure 15.2 Examples of Q_e and Q_a calculated with Mie theory for several refractive indexes.

➤ **Scattering phase matrix**

Recall definition of Stokes parameters (see Lecture 14), which uniquely characterize the electromagnetic waves. Let I_0, Q_0, U_0 and V_0 be the Stokes parameters of incident field and I, Q, U and V be the Stokes parameters of scattered radiation

$$\begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \frac{\sigma_s}{4\pi R^2} P \begin{bmatrix} I_0 \\ Q_0 \\ U_0 \\ V_0 \end{bmatrix} \quad [15.18]$$

where P is the **scattering phase matrix**.

$$P = \begin{bmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{12} & P_{22} & 0 & 0 \\ 0 & 0 & P_{33} & -P_{34} \\ 0 & 0 & P_{34} & P_{44} \end{bmatrix} \quad [15.19]$$

where each element depends on the scattering angle ($1/R^2$ is from solid angle)

For spheres: $P_{22} = P_{11}$ and $P_{44} = P_{33}$

NOTE: In general, for a particle of any shape, the scattering phase matrix consists of 16 independent elements, but for a sphere this number reduces to four.

Thus **for spheres**, Eq.[15.18] reduces to

$$\begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \frac{\sigma_s}{4\pi R^2} \begin{bmatrix} P_{11} & P_{12} & & \\ P_{12} & P_{11} & & \\ & & P_{33} & -P_{34} \\ & & P_{34} & P_{33} \end{bmatrix} \begin{bmatrix} I_0 \\ Q_0 \\ U_0 \\ V_0 \end{bmatrix} \quad [15.20]$$

where each element of the scattering phase matrix is expressed via the scattering amplitudes $S_1(\Theta)$ and $S_2(\Theta)$

$$P_{11} = \frac{4\pi}{2k^2\sigma_s} [S_1 S_1^* + S_2 S_2^*]$$

$$P_{12} = \frac{4\pi}{2k^2\sigma_s} [S_2 S_2^* - S_1 S_1^*] \quad [15.21]$$

$$P_{33} = \frac{4\pi}{2k^2\sigma_s} [S_2 S_1^* + S_1 S_2^*]$$

$$-P_{34} = \frac{4\pi}{2k^2\sigma_s} [S_1 S_2^* - S_2 S_1^*]$$

$P_{11}(\Theta) = P(\Theta)$ is the scattering phase function defined in Lecture 14.

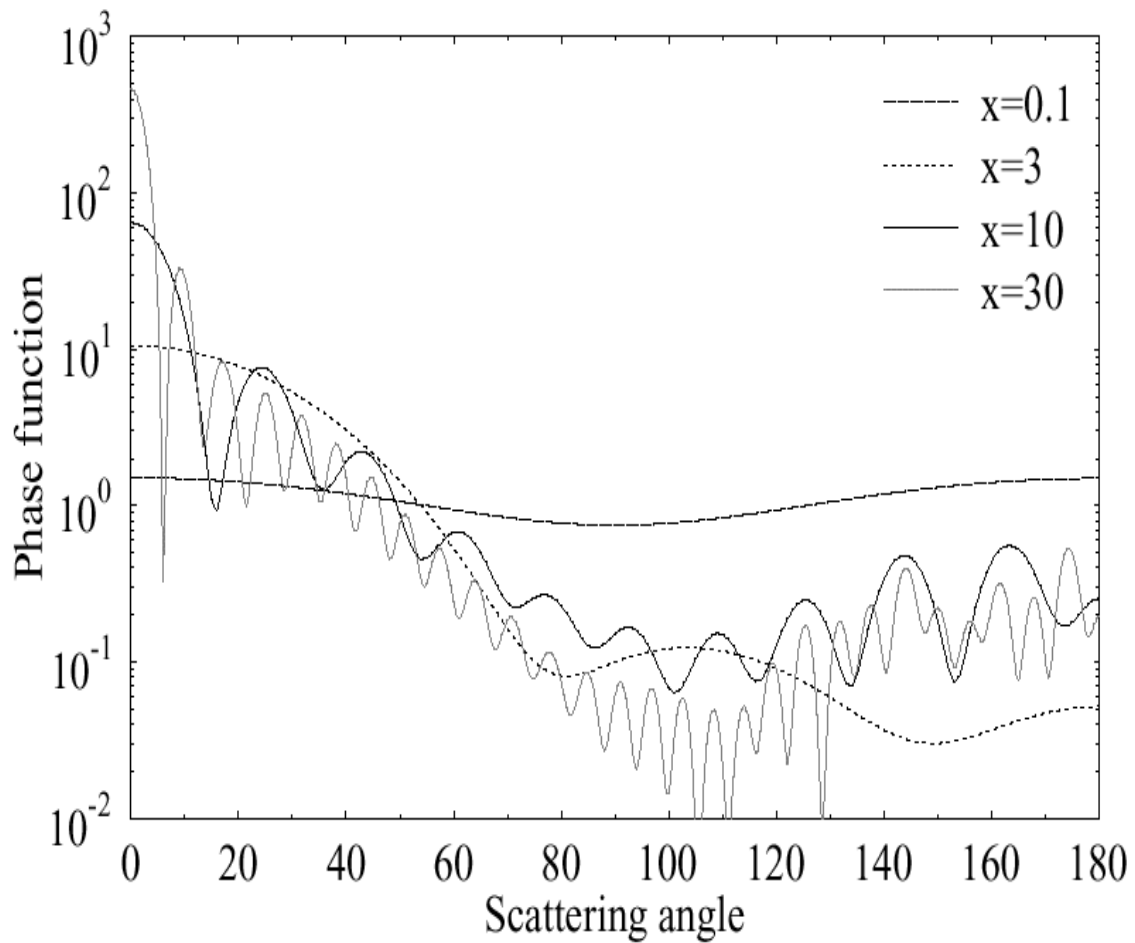


Figure 15.3 Examples of scattering phase functions calculated with Mie theory for several size parameter for nonabsorbing spheres. Note increasing oscillating behavior with increasing size parameter.

Some highlights of Mie scattering results:

- Extinction efficiency vs. size parameter x (assuming NO ABSORPTION):
 - 1) small in Rayleigh limit: $Q_e \propto x^4$
 - 2) largest Q_e when particles and wavelength have similar size
 - 3) $Q_e \rightarrow 2$ in the geometric limit ($x \rightarrow \infty$)
 - 4) Oscillations (see Fig.15.3) from interference of transmitted and diffracted waves
- Period in x of interference oscillations depends on the refractive index. Absorption reduces interference oscillations and kills ripple structure.
- Scattering and absorption efficiencies vs. size parameter with ABSORPTION:
As $x \rightarrow \infty$: $Q_s \rightarrow 1$ and , entering rays are absorbed inside particle. Smaller imaginary part of the refractive index requires larger particle to fully absorb internal rays.
- Scattering phase function: forward peak height increases dramatically with x . For single particles – number of oscillations in $P(\Theta)$ increases with x .

3. Volume optical properties of an ensemble of particles.

- ✓ Mie theory gives the extinction, scattering and absorption cross-sections (and efficiencies) and the scattering phase matrix of a single spherical particle.

NOTE: Recall Lecture 4 where the particle size distributions were introduced for atmospheric aerosols and clouds.

If the particles characterized by a size distribution $N(r)$, the volume extinction, scattering and absorption coefficients (in units LENGTH^{-1}) are calculated as

$$\begin{aligned}\beta_e &= \int_{r_{\min}}^{r_{\max}} \sigma_e(r) N(r) dr \\ \beta_s &= \int_{r_{\min}}^{r_{\max}} \sigma_s(r) N(r) dr \\ \beta_a &= \int_{r_{\min}}^{r_{\max}} \sigma_a(r) N(r) dr\end{aligned}\quad [15.22]$$

where σ is the corresponding cross section of a particle of radius r and $N(r)$ is the particle size distribution (e.g., in units $\text{m}^{-3}\mu\text{m}^{-1}$).

Single scattering albedo (unitless) is defined as

$$\omega_0 = \frac{\beta_s}{\beta_e} \quad [15.23]$$

- ✓ The **single scattering albedo** gives the percentage of light which will be scattered in a single scattered event.

Scattering phase function is

$$P(\Theta) = \frac{4\pi}{2k^2\beta_s} \int_{r_{\min}}^{r_{\max}} [S_1 S_1^* + S_2 S_2^*] N(r) dr \quad [15.24]$$

or

$$P(\Theta) = \frac{\int_{r_{\min}}^{r_{\max}} P_r(\Theta) \sigma_s N(r) dr}{\beta_s} \quad [15.25]$$

Asymmetry parameter is defined as the first moment of the scattering phase function

$$g = \frac{1}{2} \int_{-1}^1 P(\cos \Theta) \cos(\Theta) d(\cos \Theta) \quad [15.26]$$

$g = 0$ for equal forward and backward scattering; $g = 1$ for totally forward scattering

The Henyey-Greenstein scattering phase function is a model phase function, which is often used in radiative transfer calculations to approximate aerosol scattering:

$$P_{HG}(\Theta) = \frac{1 - g^2}{(1 + g^2 - 2g \cos \Theta)^{3/2}} \quad [15.27]$$

where g is the asymmetry parameter.

➤ **Optical properties of cloud drops:**

- For many practical applications, the optical properties of water clouds are parameterized as a function of the **effective radius** and **liquid water content** (LWC).

The **effective radius** is defined as

$$r_e = \frac{\int r^3 N(r) dr}{\int r^2 N(r) dr} \quad [15.28]$$

where $N(r)$ is the particle size distribution (e.g., in units $\text{m}^{-3}\mu\text{m}^{-1}$).

The **liquid water content** (LWC) was defined in Lecture 4 (see Eq.[4.7]):

$$LWC = \rho_w V = \frac{4}{3} \rho_w \int \pi r^3 N(r) dr \quad [14.29]$$

Using that the extinction coefficient of cloud droplets is

$$\beta_e = \int \sigma_e(r) N(r) dr = \int Q_e \pi r^2 N(r) dr$$

and that $Q_e \approx 2$ for water droplets at solar wavelengths, we have

$$\beta_e \approx \frac{3}{2} \frac{LWC}{r_e \rho_w} \quad [14.30]$$

➤ **Effective optical properties of an atmospheric layer consisting of gas, and aerosol and/or cloud particles:**

Effective (also called total) optical depth:

$$\tau_\lambda = \tau_{a,\lambda}^M + \tau_{s,\lambda}^M + \tau_{a,\lambda}^A + \tau_{s,\lambda}^A \quad [15.31]$$

where $\tau_{a,\lambda}^M$ and $\tau_{s,\lambda}^M$ are optical depth due to absorption by gases and molecular (Rayleigh) scattering, respectively;

$\tau_{a,\lambda}^A$ and $\tau_{s,\lambda}^A$ are optical depth due to absorption and scattering by aerosol (and/or cloud) particles, respectively.

Effective single scattering albedo:

$$\omega_{0,\lambda} = \frac{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A}{\tau_\lambda} \quad [15.32]$$

Effective scattering phase function:

$$P_\lambda(\Theta) = \frac{\tau_{s,\lambda}^M P_\lambda^M(\Theta) + \tau_{s,\lambda}^A P_\lambda^A(\Theta)}{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A} \quad [15.33]$$

Effective asymmetry parameter:

$$g_\lambda = \frac{\tau_{s,\lambda}^A g_\lambda^A}{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A} \quad [15.34]$$