Lecture 9

Rayleigh scattering.

Scattering and absorption by aerosols and clouds.

Objectives:
1. Principles of scattering.
2. Rayleigh scattering.
3. Scattering and absorption by aerosol and cloud particles.

Required Reading:
G: 5.1-5.4, 5.6, 5.7

Additional/advanced Reading:
G: 5.5,

1. Principles of scattering.

Consider a single arbitrary particle consisted of many individual dipoles. The incident electromagnetic field induces dipole oscillations. The dipoles oscillate at the frequency of the incident field and therefore scatter radiation in all directions. In a given direction of observation, the total scattered field is a superposition of the scattered wavelets of these dipoles, accounting for their phase difference: scattering by the dipoles is coherent (i.e., there is a definite relation between phases).
**Scattering phase function** $P(\cos \Theta)$ is defined as a non-dimensional parameter to describe the angular distribution of the scattered radiation as

$$\frac{1}{4\pi} \int_0^\pi P(\cos \Theta) \, d\Omega = 1 \quad [9.1]$$

where $\Theta$ is called the **scattering angle** between the direction of incidence and observation.

**NOTE:** Another form of [9.1]

$$\frac{1}{4\pi} \int_0^{2\pi} \int_0^\pi P(\cos \Theta) \sin \Theta \, d\Theta \, d\varphi = 1 \quad [9.2]$$

**NOTE:** The phase function is often expressed as

$$P(\cos \Theta) = P(\theta', \varphi', \theta, \varphi')$$

where $(\theta', \varphi')$ and $(\theta, \varphi)$ are the spherical coordinates of incident beam and direction of observation. Using the spherical geometry it can be shown (see G: Appendix 1) that

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

The **asymmetry factor** $g$ is defined as

$$g = \frac{1}{2} \int_{-1}^{1} P(\cos \Theta) \cos \Theta d(\cos \Theta) \quad [9.3]$$

$g=0$ for isotropic scattering

**Forward scattering** refers to the observations directions for which $\Theta < \pi/2$:

$g>0$ scattering in the forward direction

**Backward scattering** refers to the observations directions for which $\Theta > \pi/2$:

$g<0$ scattering in the backward direction

**NOTE:** $\Theta = 0^\circ$ is often called forward scattering and $\Theta = 180^\circ$ is called backscattering.
Scattering domains:

Rayleigh scattering: \(2\pi r/\lambda << 1\), and the refractive index \(m\) is arbitrary (applies to scattering by molecules and small aerosol particles)

Rayleigh-Gans scattering: \((m - 1) << 1\) (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)

Geometrical optics: \(2\pi r/\lambda\) is very large and \(m\) is real (applies to scattering by large cloud droplets and ice crystals).

2. Rayleigh scattering

- Because the sizes of atmospheric molecules are much smaller than the wavelengths of solar and IR radiation, the scattering by atmospheric gases is the Rayleigh scattering.
- In the Rayleigh scattering approximation, a molecule (or a small particle) is considered as an individual dipole.

Consider a small homogeneous spherical particle (e.g., a molecule) with size smaller than the wavelength of incident radiation \(\bar{E}_0\). Let \(\vec{p}_0\) be the induced dipole moment, then from the classical electromagnetic theory we have

\[
\vec{p}_0 = \alpha \bar{E}_0
\]

where \(\alpha\) is the polarizability of the particle.

NOTE: Do not confuse the polarization of the medium with polarization associated with the EM wave.

The scattered electric field at the large distance \(r\) (called far field scattering) from the dipole is given (in cgs units) by

\[
\bar{E} = \frac{1}{c^2} \frac{1}{r} \frac{\partial \vec{p}}{\partial t} \sin(\gamma)
\]
where \( \gamma \) is the angle between the scattered dipole moment \( \vec{p} \) and the direction of observation. In oscillating periodic field, the dipole moment is given in terms of induced dipole moment by

\[
\vec{p} = \vec{p}_0 \exp(-ik(r-ct))
\]  

and thus the electrical field is

\[
\vec{E} = -\vec{E}_0 \frac{\exp(-ik(r-ct))}{r} k^2 \alpha \sin(\gamma)
\]

Decomposing the electrical vector on two orthogonal components perpendicular and parallel to the plane of scattering (a plane containing the incident and scattering beams), we have

\[
E_r = -E_{0r} \frac{\exp(-ik(r-ct))}{r} k^2 \alpha \sin(\gamma_1)
\]

\[
E_i = -E_{0i} \frac{\exp(-ik(r-ct))}{r} k^2 \alpha \sin(\gamma_2)
\]

Using that

\[
I = \frac{1}{\Delta \Omega} \frac{c}{4\pi} |E|^2
\]

the perpendicular and parallel intensities (or linear polarized intensities) are

\[
I_r = I_{0r} k^4 \alpha^2 / r^2
\]

\[
I_i = I_{0i} k^4 \alpha^2 \cos^2(\Theta) / r^2
\]
Using that the natural light (incident beam) in not polarized \((I_{0r} = I_{0l} = I_0/2)\) and that 
\(k = 2\pi/\lambda\), we have

\[
I = I_r + I_l = \frac{I_0}{r^2} \alpha^2 \left( \frac{2\pi}{\lambda} \right)^4 \left( 1 + \cos^2 (\Theta) \right) \frac{1}{2}
\]

[9.13]

Eq.[ 9.13] gives the intensity scattered by molecules (Rayleigh scattering) for unpolarized incident light.

**Rayleigh scattering phase function** for incident unpolarized radiation is

\[
P(\cos(\Theta)) = \frac{3}{4} (1 + \cos^2 (\Theta))
\]

[9.14]

Eq.[ 9.13] may be rewritten in the form

\[
I(\cos(\Theta)) = \frac{I_0}{r^2} \alpha^2 \frac{128\pi^5}{3\lambda^4} P(\Theta) \frac{P(\Theta)}{4\pi}
\]

[9.15]

Eq. [9.15] may be rewritten in the terms of the scattering cross section

\[
I(\cos(\Theta)) = \frac{I_0}{r^2} \sigma_s \frac{P(\Theta)}{4\pi}
\]

[9.16]

Here the scattering cross section (in units or area) by a single molecule is

\[
\sigma_s = \alpha^2 \frac{128 \pi^5}{3\lambda^4}
\]

[9.17]

The **polarizability** is given by the Lorentz-Lorenz formula

\[
\alpha = \frac{3}{4\pi N_s} \left( \frac{m^2 - 1}{m^2 + 2} \right)
\]

[9.18]

where \(N_s\) is the number of molecules per unit volume and \(m = n - ik\) in the refractive index.

For air molecules in solar spectrum: \(n\) is about 1 but depends on \(\lambda\), and \(k = 0\).

Thus the polarizability can be approximated as

\[
\alpha = \frac{1}{4\pi N_s} \left( n^2 - 1 \right)
\]

[9.19]
Therefore, the scattering cross section of an air molecule becomes

\[ \sigma_s = \frac{8 \pi^3 (n^2 - 1)^2}{3 \lambda^4 N_s^2} f(\delta) \]

where \( f(\delta) \) is the correction factor for the anisotropic properties of air molecules, defined as \( f(\delta) = (6 + 3\delta)/(6 - 7\delta) \) and \( \delta = 0.035 \)

Using this scattering cross section, one can calculate the optical depth of the entire atmosphere due to molecular scattering as

\[ \tau(\lambda) = \sigma_s(\lambda) \int_0^{top} N(z) \, dz \]

NOTE: The Rayleigh scattering cross section and hence optical depth are inversely proportional to the fourth power of the wavelength (⇒ blue color of the sky)

Approximation of molecular Rayleigh optical depth (i.e., optical depth due to molecular scattering) down to pressure level \( p \) in the Earth’s atmosphere:

\[ \tau(\lambda) = 0.0088 \left( \frac{p}{1013 \text{ mb}} \right) \lambda^{-4.15 + 0.2 \lambda} \]

- Rayleigh scattering results in the sky polarization. The degree of linear polarization is

\[ LP(\Theta) = - \frac{Q}{I} = - \frac{I_I - I_r}{I_I + I_r} = \frac{\cos^2 \Theta - 1}{\cos^2 \Theta + 1} = \frac{\sin^2 \Theta}{\cos^2 \Theta + 1} \]
3. Scattering and absorption by aerosol and cloud particles: 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;

ii) Particle is homogeneous (therefore it is characterized by a single refractive index \( m = n - ik \) at a given wavelength);

NOTE: Mie theory requires the relative refractive index = refractive index of a particle/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.

- Mie theory calculates the scattered 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 particle).

In the far-field zone (i.e., at the large distances \( r \) from a sphere), the solution of the vector wave equation can be obtained as

\[
\begin{bmatrix}
E_i^s \\
E_r^s
\end{bmatrix} = \frac{\exp(-ikr + ikz)}{ikr} \begin{bmatrix}
S_2 & S_3 \\
S_4 & S_1
\end{bmatrix} \begin{bmatrix}
E_i^i \\
E_r^i
\end{bmatrix}
\]  

[9.24]

here \( k = \frac{2\pi}{\lambda} \), \( E_i^i \) and \( E_r^i \) are the parallel and perpendicular components of incident electrical field, and \( E_i^s \) and \( E_r^s \) are the parallel and perpendicular components of scattered electrical 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: \(S_3(\Theta) = S_4(\Theta) = 0\), and thus Eq.[9.24] gives
\[
\begin{bmatrix}
E_i^s \\
E_r^s
\end{bmatrix} = \exp(-ikr + ikz) \begin{bmatrix} S_2(\Theta) & 0 \\ 0 & S_1(\Theta) \end{bmatrix} \begin{bmatrix} E_i^i \\
E_r^i
\end{bmatrix}
\] [9.25]

Eq.[9.25] is a fundamental equation of scattered radiation by a sphere including polarization.

Mie theory scattering amplitudes:
\[
S_1(\Theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[a_n \pi_n(\cos \Theta) + b_n \tau_n(\cos \Theta)\right]
\] [9.26]
\[
S_2(\Theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[b_n \pi_n(\cos \Theta) + a_n \tau_n(\cos \Theta)\right]
\] [9.27]

where \(\pi_n\) and \(\tau_n\) are Mie angular functions
\[
\pi_n(\cos \Theta) = \frac{1}{\sin(\Theta)} P_n^1(\cos \Theta)
\] [9.28]
\[
\tau_n(\cos \Theta) = \frac{d}{d\Theta} P_n^1(\cos \Theta)
\] [9.29]

where \(P_n^1\) are the associated Legendre polynomials.

Mie theory also gives the scattering phase matrix \(P(\Theta)\) that relates the Stokes parameters \(\{I_0, Q_0, U_0, V_0\}\) of incident radiation field and the Stokes parameters \(\{I, Q, U, V\}\) of scattered radiation
\[
\begin{bmatrix}
I \\
Q \\
U \\
V
\end{bmatrix} = \frac{\sigma_s}{4\pi r^2} P \begin{bmatrix} I_0 \\
Q_0 \\
U_0 \\
V_0
\end{bmatrix}
\] [9.30]
\[
P = \begin{bmatrix}
P_{11} & P_{12} & 0 & 0 \\
P_{12} & P_{22} & 0 & 0 \\
0 & 0 & P_{33} & -P_{34} \\
0 & 0 & P_{34} & P_{44}
\end{bmatrix}
\]

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.[9.30] reduces to

\[
\begin{bmatrix}
I \\
Q \\
U \\
V
\end{bmatrix}
= \frac{\sigma_s}{4\pi r^2}
\begin{bmatrix}
P_{11} & P_{12} & 0 & 0 \\
P_{12} & P_{11} & 0 & 0 \\
0 & 0 & P_{33} & -P_{34} \\
0 & 0 & P_{34} & P_{33}
\end{bmatrix}
\begin{bmatrix}
I_o \\
Q_o \\
U_o \\
V_o
\end{bmatrix}
\]

[9.32]

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

\( P_{11}(\theta) = P(\theta) \) is the scattering phase function of a particle.

From Mie theory it follows that the extinction cross-section of a particle is

\[
\sigma_e = \frac{4\pi}{k^2} \text{Re}[S(0^0)]
\]

[9.33]

But for the forward direction (i.e. \( \Theta = 0^0 \)) from Eqs.[9.26]-9.27], we have

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

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

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

[9.34]

where \( \pi r^2 \) is the particle area projected onto the plane perpendicular to the incident beam.
Mie efficiency factors are derived from the Mie scattering amplitude

\[ Q_e = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n + 1) \text{Re} \left[ a_n + b_n \right] \]  

[9.35]

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

[9.36]

and the absorption efficiency can be calculated as

\[ Q_a = Q_e - Q_s \]  

[9.37]

- 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 geometric limit (\( x \rightarrow \infty \))

Figure 9.1 Examples of \( Q_e \) calculated with Mie theory for several refractive indexes.

Thus, for a single spherical particle Mie theory gives the extinction, scattering and absorption cross-sections and efficiency factors, the scattering amplitudes and phase matrix.
**Strategy to compute optics of an ensemble of spherical particles:**

Particle size distribution, $N(r)$;  
Refractive index, $m(\lambda)$

---

**Integration over size distribution $N(r)$**

- Scattering cross section, $\sigma_s$  
- Absorption cross section, $\sigma_a$  
- Extinction cross section, $\sigma_e$  
- Phase function, $P_{II}(\Theta)$  
  (as a function of a particle size and wavelength)

---

**Integration over the size distribution**

For a given type of particles characterized by the size distribution $N(r)$, the extinction, scattering and absorption coefficients (in units LENGTH$^{-1}$) are determined as

$$k_e = \int_{r_i}^{r_f} \sigma_e (r) N(r) dr = \int_{r_i}^{r_f} \pi r^2 Q_e N(r) dr$$  \[9.38\]

$$k_s = \int_{r_i}^{r_f} \sigma_s (r) N(r) dr = \int_{r_i}^{r_f} \pi r^2 Q_s N(r) dr$$  \[9.39\]

$$k_a = \int_{r_i}^{r_f} \sigma_a (r) N(r) dr = \int_{r_i}^{r_f} \pi r^2 Q_a N(r) dr$$  \[9.40\]
The **single scattering albedo** gives the percentage of light which will be scattered in a single scattered event and it is defined as

\[ \omega_0 = \frac{k_s}{k_e} \]  

[9.41]

**NOTE:** No absorption (conservative scattering): \( \omega_0 = 1 \)

No scattering: \( \omega_0 = 0 \)

**Scattering phase function** of particles characterized by the size distribution \( N(r) \)

\[
P(\Theta) = \frac{\int_{r_1}^{r_2} P(\Theta, r) \sigma_s N(r) dr}{\int_{r_1}^{r_2} \sigma_s N(r) dr}
\]  

[9.42]

Figure 9.2  Examples of scattering phase functions at a wavelength of 0.5 \( \mu \text{m} \).
Optical properties of the external mixture (i.e., the mixture of several types of particles)

\[ k_e = \sum_i k^i_e \quad k_s = \sum_i k^i_s \quad k_a = \sum_i k^i_a \quad [9.43] \]

where \( k^i_e \), \( k^i_s \) and \( k^i_a \) are calculated for each particle type characterized by its particle size distribution \( N_i(r) \) and a refractive index (or effective refractive index) \( m_i \).

NOTE: Do not sum the single scattering albedo and scattering phase functions !!!

- In general, an atmospheric layer has molecules, aerosols and/or cloud particles. Thus, one needs to calculate the effective optical properties of this layer as an external mixture of the optical properties of these constituents.

How to calculate the effective optical properties of an atmospheric layer consisting of gas and aerosols (or clouds):

Effective (also called total) optical depth:

\[ \tau_\lambda = \tau_{s,\lambda}^M + \tau_{a,\lambda}^M + \tau_{s,\lambda}^A + \tau_{a,\lambda}^A \quad [9.44] \]

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 particles, respectively.

Effective single scattering albedo:

\[ \omega_{0,\lambda} = \frac{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A}{\tau_\lambda} \quad [9.45] \]

Effective scattering phase function:

\[ P_\lambda(\Theta) = \frac{\tau_{s,\lambda}^M P_{s,\lambda}^M(\Theta) + \tau_{s,\lambda}^A P_{s,\lambda}^A(\Theta)}{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A} \quad [9.46] \]
Effective asymmetry parameter:

\[ g_{\lambda} = \frac{\tau_{s,\lambda}^M g_{\lambda}^A}{\tau_{s,\lambda} + \tau_{s,\lambda}^A} \]  \[9.47\]

➢ **Optics of cloud particles**

- In contrast to atmospheric aerosols, the cloud droplets consist of a single species – water. Thus, to calculate the absorption and scattering cross sections of cloud droplets, one needs to know the size of droplets and the refractive index of water versus wavelength.

**NOTE**: Cloud droplet sizes vary from a few micrometers to 100 micrometers with average diameter in 10 to 20 µm range.

Using the **effective radius** (see lecture 8)

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

and the **liquid water content** (LWC) (see Lecture 8):

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

the extinction coefficient of cloud droplets is

\[ k_e = \int \sigma_e(r)N(r)dr = \int Q_e \pi r^2 N(r)dr \]

and that \( Q_e = 2 \) for water droplets at solar wavelengths, we have

\[ k_e \approx \frac{3}{2} \frac{LWC}{r_e \rho_w} \] \[9.48\]
Figure 9.3  Example of optical properties of typical cumulus and stratus clouds (for a cloud droplet size distribution $r_{\text{eff}} = 6 \, \mu m$). Here the normalized extinction coefficient is $k_e(\lambda) / k_e(0.5 \, \mu m)$ and $k_e(0.5 \, \mu m) = 42.8 \, km^{-1}$. 
Differences in optics of water drops and ice particles are due to

i) Particle shapes;

ii) Differences between the refractive indices of water and ice.

**Approaches to predict ice crystal optics:**

i) Ray-optics approach (also called ray tracing approach) based on geometrical optics for the size parameters \((2\pi r/\lambda)\) of 15-20 and larger;

ii) Finite differences time domain (FDTD) method for the size parameters smaller than about 20;

iii) Laboratory measurements of optical properties (e.g., phase function) of the ice crystals of known shapes and sizes.