Atmospheric Aerosol

A special topic in environmental physics

Cornelia Schlundt
cornelia@iup.physik.uni-bremen.de
http://www.iup.uni-bremen.de/~cornelia/

Institut für Umweltphysik und Fernerkundung,
Universität Bremen

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Bibliography



C. F. Bohren and D. R. Huffman.

Absorption and Scattering of Light by Small Particles, ISBN: 0-471-29340-7, *Wiley-VCH*, 1998.



K. N. Liou.

An introduction to atmospheric radiation, International geophysics series, Vol. 84, *Academic Press. Second edition*, ISBN: 0-12-451451-0, 2002.



C. F. Bohren and E. Clothiaux.

Fundamentals of Atmospheric Radiation: An Introduction with 400 Problems, ISBN: 3-527-40503-8, *Wiley-VCH*, 2006.



J. P. Burrows, P. Borrell, and U. Platt (Eds.).

The Remote Sensing of Tropospheric Composition from Space, Series: Physics of Earth and Space Environments, doi:10.1007/978-3-642-14791-3, *Springer*, 2011.

List of symbols

Symbol	Name
$\tilde{\varepsilon}$; α_a ; α_s	extinction; absorption; scattering **coefficient**
ε ; κ ; σ	extinction; absorption; scattering **cross section**
Q_e ; Q_a ; Q_s	extinction; absorption; scattering **efficiency**
$\tau_\lambda(z) = \int_z^{z_\infty} \tilde{\varepsilon}_\lambda(z')dz'$	optical thickness (depth)
$\omega_0 = \alpha_s/\tilde{\varepsilon}$	single scattering albedo
$x = 2\pi r/\lambda$	size parameter
$m = m_r + i\,m_i$	complex refractive index
P	scattering phase function
θ	zenith angle (scattering angle)
ϕ	azimuth angle
L_λ	radiance (intensity)
g	asymmetry parameter

Part III

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Scattering

- State of a plane electromagnetic wave defined by
  - wavelength λ,
  - wave vector $\underline{\mathbf{k}}$,
  - vectors of electric fields $\underline{\mathbf{E}}_1 = E_1\hat{\underline{\mathbf{e}}}_1$, $\underline{\mathbf{E}}_1 = E_2\hat{\underline{\mathbf{e}}}_2$
  - perpendicular to the direction of propagation of the wave $\underline{\mathbf{k}}/k$

$$\underline{\mathbf{E}}(\underline{\mathbf{r}}, t) = (E_1\hat{\underline{\mathbf{e}}}_1 + E_2\hat{\underline{\mathbf{e}}}_2)e^{i\underline{\mathbf{k}}\cdot\underline{\mathbf{r}} - i\omega t}$$

- Complex field amplitudes $E_1 = |E_1|e^{i\delta_1}$ and $E_2 = |E_2|e^{i\delta_2}$ describe the state of polarization of the wave

Stokes vector

- Stokes vector $\underline{\mathbf{I}} = (I, Q, U, V)$

$$\begin{aligned} I &= |E_1|^2 + |E_2|^2 \\ Q &= |E_1|^2 - |E_2|^2 \\ U &= 2|E_1||E_2|\cos\delta \\ V &= 2|E_1||E_2|\sin\delta \end{aligned}$$

- Phase difference: $\delta = \delta_2 - \delta_1$
- $I \rightarrow$ intensity of the wave
- $Q, U, V \rightarrow$ polarization configuration

Scattered Stokes vector

- Scattered radiation field
  $\rightarrow$ general description of the scattered Stokes vector $\underline{\mathbf{I}}$ in the far-field region (i. e., at a distance R to the particle $\gg \lambda$)

$$\underline{\mathbf{I}} = \alpha_s \underline{\underline{\mathbf{P}}} \cdot \underline{\mathbf{I_0}} \frac{d\upsilon}{4\pi R^2}$$

$\underline{\mathbf{I_0}}$	...	Stokes vector of the incident radiation
$d\upsilon$	...	small volume element where the scattering process takes place
α_s	...	scattering coefficient [length^{-1}]
$\underline{\underline{\mathbf{P}}}$	...	4×4 scattering matrix (Mueller matrix)

- Scattering coefficient is equal to the scattering cross section σ [area] per unit volume:

$$\alpha_s = \frac{\sigma}{d\upsilon} = \frac{1}{d\upsilon}\sum_i \sigma_i$$

- Scattering efficiency defined as dimensionless ratio of scattering cross section to the particle cross-sectional area projected onto a plane perpendicular to the incident beam
(e. g., $G = \pi a^2$ for a sphere of radius a):

$$Q_{s,i} = \frac{\sigma_i}{G_i}$$

- Scattering matrix $\underline{\underline{\mathbf{P}}}(\theta)$ describes the angular distribution of the intensity and polarization of the scattered radiation.

  In other words:
  Mueller matrix describes the relation between “incident” and “transmitted” Stokes vectors;

  - by “incident” is meant before interaction with the optical element
  - by “transmitted” is meant after interaction

- Scattering phase function: $P(\theta) = P_{11}(\theta)$
It gives the probabilbity of scattering of a light photon into the direction of the scattering angle θ.
- Polarized phase function: $Q(\theta) = P_{12}(\theta)$
or phase function of degree of polarization
It gives the degree of polarization for a scattering into the direction θ.

- Overall probabilbity for the scattering process equals 1
  $\Rightarrow$ **phase function is normalized** to one:

$$\int_{4\pi} = \frac{P_{11}(\theta)}{4\pi} d\Omega = 1$$

  $d\Omega \cdots$ unit solid angle

- Asymmetry parameter g describes the anisotropy of the scattered light:

$$g = \langle \cos\theta \rangle = \int_{4\pi} \cos\theta \frac{P_{11}(\theta)}{4\pi} d\Omega$$

  $\Longrightarrow$ g between +1 and -1

$g = 1$	$\rightarrow$	forward scattering
$g = -1$	$\rightarrow$	backward scattering
$g = 0$	$\rightarrow$	isotropically scattered light (i. e., the same in all directions)

- Knowledge of $\underline{\underline{\mathbf{P}}}$ and $\alpha_s \rightarrow$ full description of scattering
- However, application in radiative transfer and remote sensing require an additional parameter describing absorption.
- Single scattering albedo ω_0 (dimensionless) defined as the ratio of scattered energy to total extinction (i. e., sum of scattered and absorbed energie)
- **Absorption cross section** κ and **absorption efficiency** Q_a
- Extinction is the sum of scattering and absorption:

$$\begin{aligned} \tilde{\varepsilon} &= \alpha_s + \alpha_a \\ \varepsilon &= \sigma + \kappa \\ Q_e &= Q_s + Q_a \end{aligned}$$

- Single scattering albedo defined as

$$\omega_0 = \frac{\alpha_s}{\tilde{\varepsilon}} = \frac{\sigma}{\varepsilon} = \frac{Q_s}{Q_e}$$

- In general, α_s, $\underline{\underline{\mathbf{P}}}$, and ω_0 depend on the orientation of the unit volume dv w.r.t. the direction of the incident light beam.
- For the most applications
  $\rightarrow \alpha_s$ and ω_0 are constant and
  $\rightarrow \underline{\underline{\mathbf{P}}}$ is a function of the scattering angle with a maximum of 6 independent matrix elements:

$$\underline{\underline{\mathbf{P}}} = \begin{pmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{21} & P_{22} & 0 & 0 \\ 0 & 0 & P_{33} & -P_{43} \\ 0 & 0 & P_{43} & P_{44} \end{pmatrix}$$

- Goal of the scattering theory $\rightarrow$ determine
  - scattering cross sections σ
  - single scattering albedo ω_0
  - scattering matrix $\underline{\underline{\mathbf{P}}}$

  for many different naturally occurring particles
- **Input parameters** in
  - radiative transfer theory
  - remote sensing
- Vice versa: **measurement of scattered radiation** in the framework of remote sensing $\longrightarrow$ information about scattering phase function $\rightarrow$ nature of particle

- Solutions for scattering problems
- Choice of appropriate method depends mainly on 3 parameters:
  1. Size parameter: $x = 2\pi r/\lambda$

     $\rightarrow$ giving the ratio of particle radius to the wavelength of the incident light
  2. Complex refractive index: $m = m_r + im_i$
  3. Particle shape

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Rayleigh scattering

- Simplest behavior of scattering is that of **small particles**.
- 1871 Lord Rayleigh established the basic characteristics
  $\rightarrow$ successful explanation of color and polarization properties of the skylight $\Rightarrow$ scattering by air molecules in the air
- **Scope of application**:
  particle size must be much smaller than the wavelength of the incident light ($x \ll 1$)
- Incident light induces a dipole moment inside of the particle, which is proportinal to the strength of the incident electric field

  The incident field oscillates as EM wave at a specific frequency
  $\rightarrow$ inducing a likewise oscillating dipole moment
  $\Rightarrow$ which in turn emits an EM field (Hertzian dipole)

- Under these conditions: radiance L of the scattered radiation in the far-field ($R \gg \lambda$) is given by:

$$L(\theta) = const\, L_0 \frac{1}{R^2} \frac{1}{\lambda^4} (1 + \cos^2(\theta))$$

θ	...	scattering angle
L_0	...	radiance of the incident radiation

- Comparing the equation above with the **definition of the scattering matrix**:

$$\underline{\mathbf{I}} = \alpha_s \underline{\underline{\mathbf{P}}} \cdot \underline{\mathbf{I_0}} \frac{d\upsilon}{4\pi R^2}$$

- One obtains for the Rayleigh-scattering function and Rayleigh-scattering coefficient:

$$P(\theta) = \frac{3}{4}(1 + \cos^2(\theta)) \qquad \alpha_s = const\, \lambda^{-4}$$

- Since λ ranges in the visible from $0.4\mu m$ (violet) to $0.8\mu m$ (red)

  $\rightarrow$ Rayleigh scattering cross section σ_λ^R is approximately 16 times larger at the blue end than at the red end

- Strong wavelength dependence of the scattering ($\sim \lambda^{-4}$)

  $\rightarrow$ shorter (blue) wavelengths are scattered more strongly than longer (red) wavelengths $\rightarrow$ blue light preferentially sscattered from light passing above (**responsible for the blue skies in the atmosphere**)

- Position of the sun is low

  $\rightarrow$ solar radiation takes a long way through the dense lower layers of the atmopshere $\rightarrow$ red light remaining in the sunlight after losses of blue scattered light (**responsible for red sunsets (sunrise)**)

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Mie scattering

- If the particle size is **not** much smaller than the wavelength of the incident light $\rightarrow$ Rayleigh scattering no longer valid
- Scattering of light by aerosols and larger particles (cloud droplets, ice crystals)
- Exact solutions for single scattering problems exist only for a few profoundly symmetric particles
- Most important exactly soluble problem in the theory of absorption and scattering by small particles $\rightarrow$ sphere of arbitrary radius and refractive index
- **Mie theory** $\rightarrow$ describing the scattering, absorption, extinction, and polarization of light at particles of spherical shape, comparable in size with the wavelength

Mie theory: absorption and scattering by a sphere

- Solutions for the scattering matrix as well as scattering and absorption cross sections consist of a converging sum of **Mie coefficients**
- Determined by **Legendre polynomials** and **shperical Bessel functions**
- Soluble using recursive formula
- Appropriate for numerical computations
- Mie theory: spherical particles, droplets
  no problems with aerosols having $r < 0.5\mu m \rightarrow x = \frac{2\pi r}{\lambda} < 5$
- For particles with $r > 10\mu m$
  $\rightarrow$ wavelength dependence disappears
  $\rightarrow$ scattering becomes gray (clouds, fog)
  $\rightarrow$ larger particles scattering mainly in forward direction

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Geometrical optics

- Light scattering at very large aspherical particles
- **Mie theory no longer valid**
- Ice crystals in cirrus clouds
- Soot and dust particles in the lower atmosphere
- Large cloud droplets
- Scattering calculations in the size range of geometrical optics can be performed by ray tracing

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- Rayleigh-Gans scattering
- Anomalous diffraction theory
- Discrete Dipole Approximation

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Other methods

- Rayleigh scattering, Mie theory, and geometrical optics cover a lot of situations that can be found in the Earth's atmosphere
- **However, there is a gap!**
  - Particle is non-spherical and its extension is larger than several wavelengths of the incident light
- Till this day: there is no exact scattering theory for abritrary shaped scattering bodies in this size range
- Several approximations do exist:
  - Rayleigh-Gans scattering
  - Anomalous diffraction theory (ADT)
  - Discrete Dipole Approximation (DDA)

Rayleigh-Gans scattering

- Generalization of Rayleigh scattering
- **Arbitrary shaped** and **medium-sized** scattering body
- Assumption: scattering particles have a refractive index ($n1$) very close to the refractive index of the surrounding medium ($n0$)
  $\rightarrow \quad m = n1/n0 \quad \rightarrow \quad |1 - m| = 1$
- Such particles can be considered as composed of very small elements
- Each of which $\rightarrow$ acting as independent Rayleigh particle
- Thus, each small element of the larger particle is assumed to scatter independently of any other.

Anomalous diffraction theory (ADT)

- Developed by van de Hulst (1957)
- Describing light scattering for large optically soft particles:
  - $x \gg 1$ and $n \rightarrow 1$
- **Rays are negligibly**
  - deviated as they cross the soft particle boundary
  - reflected because the refractive indices inside and outside the particle are similar
- In this case, **extinction is largely caused**
  - by absorption of the light beam passing through the particle
  - by the interference of light passing through the particle and passing around the particle

Anomalous diffraction theory (ADT)

- Useful for the calculation of the
  $\rightarrow$ extinction and absorption coefficients
- Can be also applied to nonspherical particles, such as :
  - spheroids
  - hexagons
- Since refractions and reflections of rays are **neglected** in this approximation, its accuracy must be examined carefully when applied to the scattering of

  $\rightarrow$ **ice crystals** ($n \sim 1.31$)

  $\rightarrow$ **aerosols** ($n \sim 1.5$)!

Discrete Dipole Approximation (DDA)

- **Basic idea**
  $\rightarrow$ introduced in 1964 by DeVoe
  - Investigation of optical properties of molecular aggregates
  - Neglecting retardation effects
  - Limited to aggregates that were small compared to the wavelength
- **DDA including retardation effects**
  $\rightarrow$ proposed in 1973 by Purcell and Pennypacker
  - Approximation of the continuum target by a finite array of polarizable points
  - Points acquire dipole moments in response to the local electric field
  - Dipoles interact with one another via their electric fields
  - DDA is referred to as the coupled dipole approximation

Discrete Dipole Approximation (DDA)

- **Arbitrary shaped** scattering body with **x** $\sim$ **1**
- Similar to Rayleigh-Gans approximation
  $\rightarrow$ particle is subdivided into sufficiently **small volume elements acting as dipoles**
- Scatter field is the result of interaction between the dipoles with one another and the incident electric field
- DDA is more accurate, the smaller the distance between the individual dipoles is compared to the wavelength,
  i. e. the greater the number of dipoles
- DDA further improved $\rightarrow$ **DDA codes available**

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7 Derivation of the Mie theory
- Morphology of atmospheric particulates
- Electromagnetic wave equation and solution
- Formal scattering solution
- The far-field solution, extinction parameters and scattering matrix

Mie theory

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Morphology of atmospheric particulates

- Earth's atmsphere contains **various types of particulates**
  - aerosols
  - water droplets
  - ice crystals
  - raindrops
  - snowflakes
  - hailstones
- Physical and dynamic processes $\rightarrow$ formation and growth of particles
- Emphasis on aerosols and cloud particles

Sources of aerosol particles

- Aerosol of natural origin:
  - dust from arid and semi-arid regions
  - particles from sea spray over the oceans
  - volcanic debris
  - smoke from forest fires
  - extraterrestrial or interplanetary dust
  - small particles produced by chemical reactions of natural gases
- Man-made aerosols produced by particles:
  - directly emitted during comustion processes
  - formed from emitted gases

Aerosol classification in terms of their location and type

- Continental type $\rightarrow$ subdivided into conditions:
  - **clear** (rural, forest)
  - **average**
  - **desert** (background, windy)
  - **urban** / **industrial**
- Aerosol components involved in this type are:
  - water-soluble particles
  - dustlike particles
  - soot
  - minerals

Aerosol classification in terms of their location and type

- Maritime type → grouped into the following conditions:
  - **clear**
  - **mineral**
  - **polluted**
- Aerosol components involved in this type are:
  - sea salt particles
  - mineral particles
  - sulfate particles
  - soot particles

Aerosol classification in terms of their location and type

- Polar type → grouped into the following conditions:
  - **polluted**
  - **clear Arctic**
  - **clear Antarctic**
- Aerosol components involved in this type are:
  - sea salt particles
  - mineral particles
  - sulfate particles
  - soot particles

- Dustlike substances:
  - mineral dust formed in nondesert locations
  - representative of soil conditions
- Water-soluble substances:
  - refer to the parts of aerosols that are soluble in water
  - consist of a mixture of sulfate, nitrate, and organic compounds
  - mixing ratio varies with the source
- Soot:
  - represents all carbonaceous materials
  - either a product of direct particle emssion into the atmsophere by combustion processes
  - or a product of the transformation of combustion-related gases to particles

- Sulfate particles:
  - products of oxidation of sulfur-bearing gases
  - generated both naturally (by biogenic sources) and anthropogenic (by coal and oil burning, smelting, petroleum refining, transportation)
- Diverse tropospheric aerosol types conveniently grouped into 5 basic categories:
  1. dustlike soil
  2. soot
  3. sulfate
  4. sea salt
  5. organic aerosols

Aerosol size distribution

- Grouped into **2 size classes**
  $\rightarrow$ representing 2 primary formation mechanisms:
  1. Particles with diameters larger than about 1 μm

     $\rightarrow$ produced by breakup and suspension ob bulk materials by the wind (e.g., sea salt and soil dust)
  2. Fine particles smaller than 1 μm

     $\rightarrow$ usually formed by combustion or chemical conversions of the gaseous precursors into liquid or solid products
- Their radiative properties strongly depend on their **refractive indices as a function of wavelength**
- In wet environments $\rightarrow$ **humidity effects important**; affecting size, shape and chemical composition
- See Part 5: Microphysics of aerosols

Microsized dust particles from the Sahara desert collected in Israel in 1998

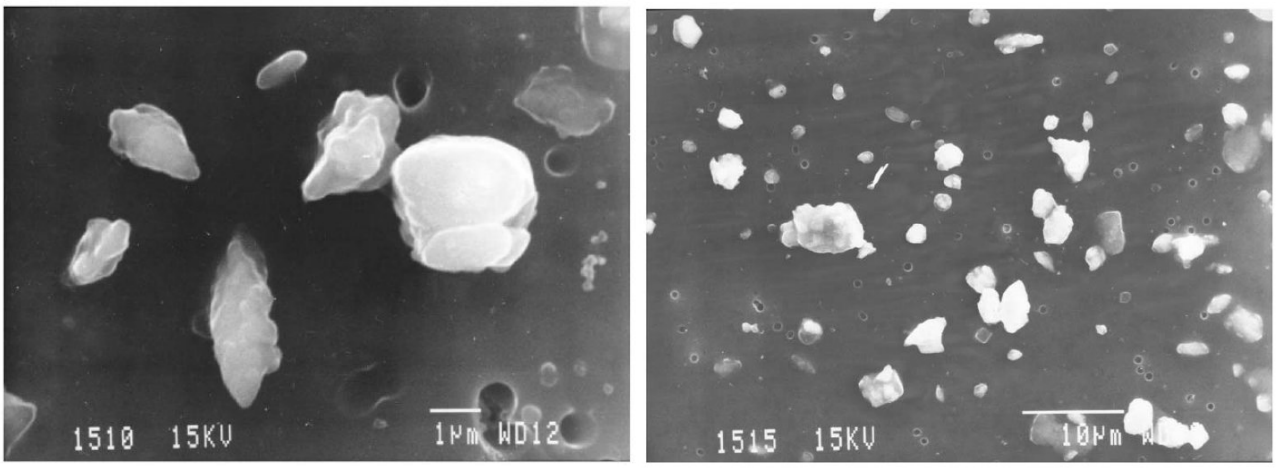


Figure 1: Note that the scales for the left and right diagrams are 1 and 10 μm, respectively.

Clouds

- Composed of water droplets and/or ice crystals
- Classified in terms of their position and appearance in the atmosphere: in midlatitudes
  - base heights of $\sim$ 6 km $\rightarrow$ **high** clouds (cirrus clouds)
  - base heights of $\sim$ 2 km $\rightarrow$ **low** clouds (stratus and cumulus)
  - between low and heigh clouds $\rightarrow$ **middle** clouds (altocumulus and altostratus)
  - significant vertical developments (e.g., in tropics) $\rightarrow$ cumulusnimbus

Clouds: microphysical state

- Determined by the dispersion of particle sizes and their phase (ice or liquid)
- Low and some middle clouds
  $\rightarrow$ generally composed of **spherical water droplets** ($\sim$ 1 μm to 20μm)
- Typical water droplet is on the order of 5 μm
- Some middle clouds with temperatures warmer than about -20°C
  $\rightarrow$ **supercooled water droplets** that coexist with ice particles
- Small water droplets are spherical in nature
  ($\rightarrow$ requirement of surface pressure to hold water molecules together)
- **Collision** and **coalescence** $\rightarrow$ mm- and cm-sized raindrops
- **Falling raindrops** in a dynamic flow field deviate from the spherical shape

Ice crystal size and shape as a function of height (in a typical midlatitude cirrus) and relative humidity

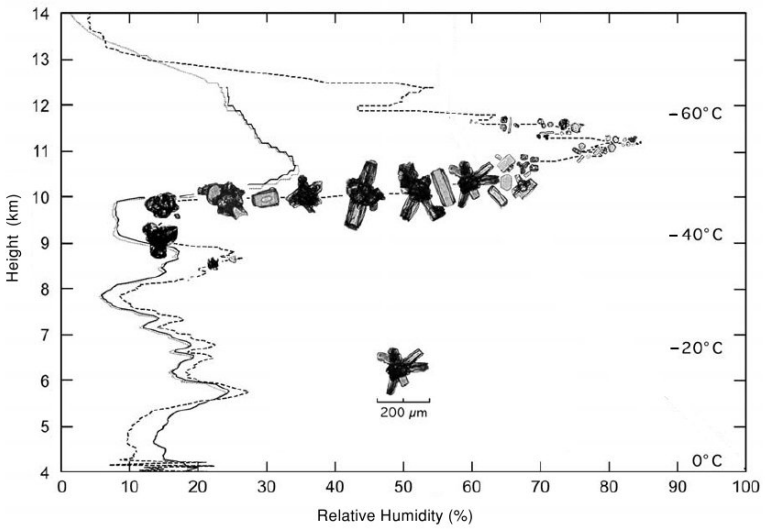


Figure 2: Measured by a replicator ballon sounding system in Marshall, Colorado, on November 10, 1994. **At the cloud top**, pristine and small columns and plates are predominant. **At the cloud bottom**, irregular bullet rosettes and aggregates are produced, presumably due to the collision and coalescence associated with vertical mixing and gravitational pulling. Ice crystals size and shape vary greatly with time and space, and thus, a presentation of representative values for the purpose of remote sensing and climate applications is a diffult task.

Maxwell equations

$$\nabla \times \vec{H} = \frac{\varepsilon}{c}\frac{\partial \vec{E}}{\partial t} \quad (1)$$

$$\nabla \times \vec{E} = -\frac{\mu}{c}\frac{\partial \vec{H}}{\partial t} \quad (2)$$

$$\nabla \cdot \vec{E} = 0 \quad (3)$$

$$\nabla \cdot \vec{H} = 0 \quad (4)$$

electric current density $\vec{j} = \sigma \vec{E}$ (5)

electric displacement $\vec{D} = \varepsilon \vec{E}$ (6)

magnetic induction $\vec{B} = \mu \vec{H}$ (7)

σ ... specific conductivity
ε ... permittivity
μ ... magnetic permeability

- **Assumption**:
  - Field $\rightarrow$ no charges ($\rho = 0$) and no currents ($|\vec{j}| = 0$) [vacuum]
  - Medium is homogeneous $\rightarrow$ ε and μ are constants
- From these 4 equations $\rightarrow$ **derivation of the EM wave equation**

EM wave equations

- Assume a **plane EM wave** in a periodic field with circular frequency ω
- Equations (8) and (9) $\rightarrow$ **electric** and **magnetic vectors** in a homogeneous medium satisfy the vector wave equation (10)
- Equation (11) $\rightarrow$ scalar wave equation for ψ

$$\nabla^2\vec{E} = -k^2m^2\vec{E} \quad (8) \qquad \nabla^2\vec{A}+k^2m^2\vec{A} = 0 \quad (10)$$

$$\nabla^2\vec{H} = -k^2m^2\vec{H} \quad (9) \qquad \nabla^2\psi+k^2m^2\psi = 0 \quad (11)$$

$k = 2\pi/\lambda = (\omega/c)$ ... wavenumber (= propagation constant in vacuum)
$m = \sqrt{\varepsilon}$ ... complex refractive index of the medium at frequency ω

- Assuming that u and v are 2 independent solutions of the scalar wave equation defined in Equation (11),
  $\rightarrow$ then the **electric and magnetic field vectors** expressed by:

$$\vec{E} = \vec{M}_v + i\vec{N}_u \quad (12)$$

$$\vec{H} = m(-\vec{M}_u + i\vec{N}_v) \quad (13)$$

- satisfy the following two equations:

$$\nabla \times \vec{H} = ikm^2\vec{E} \quad (14)$$

$$\nabla \times \vec{E} = -ik\vec{H} \quad (15)$$

- $\vec{E}$ and $\vec{H}$ can be written as follows:

$$\begin{aligned}\vec{E} &= \hat{a}_r \frac{i}{mk}\left[\frac{\partial^2(ru)}{\partial r^2} + m^2k^2(ru)\right] + \hat{a}_\theta\left[\frac{1}{r\sin\theta}\frac{\partial(rv)}{\partial\phi} + \frac{i}{mkr}\frac{\partial^2(ru)}{\partial r\,\partial\theta}\right] \\ &+ \hat{a}_\phi\left[-\frac{1}{r}\frac{\partial(rv)}{\partial\theta} + \frac{1}{mkr\sin\theta}\frac{\partial^2(ru)}{\partial r\,\partial\phi}\right] \qquad (16)\end{aligned}$$

$$\begin{aligned}\vec{H} &= \hat{a}_r \frac{i}{k}\left[\frac{\partial^2(rv)}{\partial r^2} + m^2k^2(rv)\right] + \hat{a}_\theta\left[-\frac{m}{r\sin\theta}\frac{\partial(ru)}{\partial\phi} + \frac{i}{kr}\frac{\partial^2(rv)}{\partial r\,\partial\theta}\right] \\ &+ \hat{a}_\phi\left[\frac{m}{r}\frac{\partial(ru)}{\partial\theta} + \frac{i}{kr\sin\theta}\frac{\partial^2(rv)}{\partial r\,\partial\phi}\right] \qquad (17)\end{aligned}$$

- Scalar plane wave equation (11) expressed in **polar coordinates**:

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial\psi}{\partial r}\right) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\psi}{\partial\theta}\right) + \frac{1}{r^2\sin\theta}\frac{\partial^2\psi}{\partial\phi^2} + k^2m^2\psi = 0 \qquad (18)$$

- We seek particular solutions to Equation (18) of the form:

$$\psi(r,\theta,\phi) = R(r)\,\Theta(\theta)\,\Phi(\phi) \qquad (19)$$

- The **solution** to the problem of scattering by an arbitrary homogeneous sphere illuminated by a plane wave can be obtained by expanding the incident, scattered, and internal electric and magnetic fields in series of vector spherical harmonics (Legendre functions, Bessel function, Neumann function)

$$\begin{aligned}r\psi(r,\theta,\phi) &= \sum_{n=0}^{\infty}\sum_{l=-n}^{n} P_n^l(\cos\theta) \qquad (20)\\ &\quad [c_n\psi_n(kmr) + d_n\chi_n(kmr)](a_l\cos l\phi + b_l\sin l\phi)\end{aligned}$$

Formal scattering solution

- Scattering of a plane wave by a homogeneous sphere
- For simplicity: we assume
  - outside the medium is vacuum ($m = 1$)
  - material of the sphere has an refractive index m
  - incident radiation is linearly polarized
- Origin of a rectangular system of coordinates at the center of the sphere
- Direction of propagation of the incident wave along the positive z axis
- Amplitude of the incident wave normalized to unity
- **Incident electric and magnetic field vectors** are given by:

$$\vec{E}^i = \hat{a}_x e^{-ikz} \qquad \vec{H}^i = \hat{a}_y e^{-ikz} \tag{21}$$

- Incident waves^{inc} outside the sphere:

$$\begin{aligned} ru^{inc} &= \frac{1}{k}\sum_{n=1}^{\infty}(-i)^n \frac{2n+1}{n(n+1)}\psi_n(kr)P_n^1(\cos\theta)\cos\phi \\ rv^{inc} &= \frac{1}{k}\sum_{n=1}^{\infty}(-i)^n \frac{2n+1}{n(n+1)}\psi_n(kr)P_n^1(\cos\theta)\sin\phi \end{aligned} \tag{22}$$

- In order to match u^{inc} and v^{inc} with those of the internal and scattered waves whose potentials have been derived in Equation (20), the latter must be expressed in a series of a similiar form but with arbitrary coefficients.

- For internal waves^{transmitted}, because the function $\chi_n(kmr)$ becomes infinite at the origin, only the function $\psi_n(kmr)$ may be used:

$$ru^{tra} = \frac{1}{mk}\sum_{n=1}^{\infty}(-i)^n\frac{2n+1}{n(n+1)}c_n\psi_n(kmr)P_n^1(\cos\theta)\cos\phi$$

$$rv^{tra} = \frac{1}{mk}\sum_{n=1}^{\infty}(-i)^n\frac{2n+1}{n(n+1)}d_n\psi_n(kmr)P_n^1(\cos\theta)\sin\phi \quad (23)$$

- For scattered waves^{sca}, these two functions must vanish at infinity $\rightarrow$ **Hankel functions** possess exactly this property:

$$ru^{sca} = \frac{1}{k}\sum_{n=1}^{\infty}(-i)^n\frac{2n+1}{n(n+1)}a_n\xi_n(kr)P_n^1(\cos\theta)\cos\phi$$

$$rv^{sca} = \frac{1}{k}\sum_{n=1}^{\infty}(-i)^n\frac{2n+1}{n(n+1)}b_n\xi_n(kr)P_n^1(\cos\theta)\sin\phi \quad (24)$$

- a_n and b_n are the scattering coefficients

- The coefficients a_n, b_n, c_n, and d_n must be determined from the **boundary conditions at the surface of the sphere**.
- The coefficients of these expansion functions are chosen so that the tangential components of the electric and magnetic fields are continuous across the surface of the sphere $r = a$ such that:

$$\begin{aligned} E_\theta^{inc} + E_\theta^{sca} &= E_\theta^{tra} & E_\phi^{inc} + E_\phi^{sca} &= E_\phi^{tra} \\ H_\theta^{inc} + H_\theta^{sca} &= H_\theta^{tra} & H_\phi^{inc} + H_\phi^{sca} &= H_\phi^{tra} \end{aligned} \tag{25}$$

- Thus, this scattering problem is **formally identical to reflection and refraction** because of interfaces.

- Coefficients for the **scattered** waves: with $x = ka$ and $y = mx$

$$\begin{aligned} a_n &= \frac{\psi_n'(y)\psi_n(x) - m\psi_n(y)\psi_n'(x)}{\psi_n'(y)\xi_n(x) - m\psi_n(y)\xi_n'(x)} \\ b_n &= \frac{m\psi_n'(y)\psi_n(x) - \psi_n(y)\psi_n'(x)}{m\psi_n'(y)\xi_n(x) - \psi_n(y)\xi_n'(x)} \end{aligned} \tag{26}$$

- Coefficients of the field **inside** the particle:

$$\begin{aligned} c_n &= \frac{m[\psi_n'(y)\xi_n(x) - \psi_n(y)\xi_n'(x)]}{\psi_n'(y)\xi_n(x) - m\psi_n(y)\xi_n'(x)} \\ d_n &= \frac{m[\psi_n'(y)\xi_n(x) - \psi_n(y)\xi_n'(x)]}{m\psi_n'(y)\xi_n(x) - \psi_n(y)\xi_n'(x)} \end{aligned} \tag{27}$$

- Riccati-Bessel functions with $\rho = kmr$ ($k = 2\pi/\lambda$)
  - **Bessel function of the first kind** $J_{n+1/2}$

$$\psi_n(\rho) = \sqrt{\pi\rho/2}\, J_{n+1/2}(\rho) \tag{28}$$

  - **Half-integral-order Hankel function of the second kind** $H^{(2)}_{n+1/2}$

$$\xi_n(\rho) = \psi_n(\rho) + i\chi_n(\rho) = \sqrt{\pi\rho/2}\, H^{(2)}_{n+1/2}(\rho) \tag{29}$$

- The solution of the scattering of EM waves by a sphere with a radius a and a refractive index of m is complete
- The electric and magnetic field vectors at any point inside and outside the sphere are now expressed in terms of the known mathematical functions given by Equations (22) – (24)
- So far we assumed, that the suspending medium is a vacuum
- **Outside medium**: refractive index m_2 (real part)
- **Sphere**: refractive index m_1 (maybe complex)
- By replacing m by m_1/m_2 and the wavenumber k by $m_2 k$
  $\rightarrow$ the results given in **Equation (26) can be generalized** to cases where a sphere is suspended in a medium

The far-field solution, extinction parameters and scattering matrix

- Consider the scattered field at very large distances from the sphere
- Hankel functions expressed by Equation (29) **reduce** to the form:

$$\xi_n(kr) \approx i^{n+1}\, e^{-ikr} \qquad kr \gg 1 \tag{30}$$

- With this simplification, Equations (24) become

$$\begin{aligned} ru^{sca} &\approx -\frac{ie^{-ikr}\cos\phi}{k}\sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}a_n P_n^1(\cos\theta) \\ rv^{sca} &\approx -\frac{ie^{-ikr}\sin\phi}{k}\sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}b_n P_n^1(\cos\theta) \end{aligned} \tag{31}$$

- Fields representing an outgoing spherical wave where amplitude and the state of polarization are functions of the scattering angle θ

$$\begin{aligned} E_\theta^{sca} &= E_l^{sca} = \frac{i}{kr}e^{-ikr}\cos\phi\, S_2(\theta) \\ -E_\phi^{sca} &= E_r^{sca} = \frac{i}{kr}e^{-ikr}\sin\phi\, S_1(\theta) \end{aligned} \tag{32}$$

- Scattering functions:

$$\begin{aligned} S_1(\theta) &= \sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}\left[a_n\pi_n(\cos\theta)+b_n\tau_n(\cos\theta)\right] \\ S_2(\theta) &= \sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}\left[b_n\pi_n(\cos\theta)+a_n\tau_n(\cos\theta)\right] \end{aligned} \tag{33}$$

where

$$\pi_n(\cos\theta) = \frac{1}{\sin\theta}P_n^1(\cos\theta) \qquad \tau_n(\cos\theta) = \frac{d}{d\theta}P_n^1(\cos\theta) \tag{34}$$

- **Normalized incident electric vector**
  $\rightarrow$ decomposed into perpendicular and parallel components:

$$E_r^{inc} = e^{-ikz}\sin\phi \qquad E_l^{inc} = e^{-ikz}\cos\phi \tag{35}$$

- Equation (32) can then be rewritten and the relation between incident and scattered field amplitudes is therefore:

$$\begin{bmatrix} E_l^{sca} \\ E_r^{sca} \end{bmatrix} = \frac{e^{-ikr+ikz}}{ikr} \begin{bmatrix} S_2(\theta) & 0 \\ 0 & S_1(\theta) \end{bmatrix} \begin{bmatrix} E_l^{inc} \\ E_r^{inc} \end{bmatrix} \tag{36}$$

  $\rightarrow$ representing the fundamental equation for the study of a light beam scattered by spheres including polarization!

- Computational problem involved in Mie scattering
  $\rightarrow$ evaluate the intensity functions as functions of the
  - scattering angle θ
  - index of refraction m
  - particle size parameter $x = 2\pi r/\lambda$

- Intensity functions:

$$i_1(\theta) = |S_1(\theta)|^2 \qquad i_2(\theta) = |S_2(\theta)|^2 \tag{37}$$

- In the far-field zone $\rightarrow$ evaluation of the reduction of the incident energy due to the absorption and scattering of light by a sphere

- In the forward direction $\rightarrow \theta \approx 0$
- Extinction cross section:

$$\varepsilon = \left(\frac{4\pi}{k^2}\right) Re[S(0)] \tag{38}$$

- Extinction efficiency of a sphere
  with a radius of $r = a$ and $x = ka$ (size parameter):

$$Q_e = \frac{\varepsilon}{\pi a^2} = \frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)Re(a_n + b_n) \tag{39}$$

- Scattering cross section:

$$\sigma = \frac{\text{total flux of scattered light}}{\text{incident flux density}} = \frac{\pi}{k^2}\int_0^{\pi}[i_1(\theta) + i_2(\theta)]\sin\theta d\theta \tag{40}$$

- Scattering efficiency:

$$Q_s = \frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)(|a_n|^2 + |b_n|^2) \tag{41}$$

- Based on **energy conservation principle**
  $\rightarrow$ absorption cross section and efficiency of a sphere
  can be derived from the following relationships:

$$\kappa = \varepsilon - \sigma \qquad Q_a = Q_e - Q_s \tag{42}$$

- For an absorbing sphere
  $\rightarrow$ convenient to define a complex refractive index: $m = m_r - im_i$

Scattering efficiency factor as a function of the size parameter

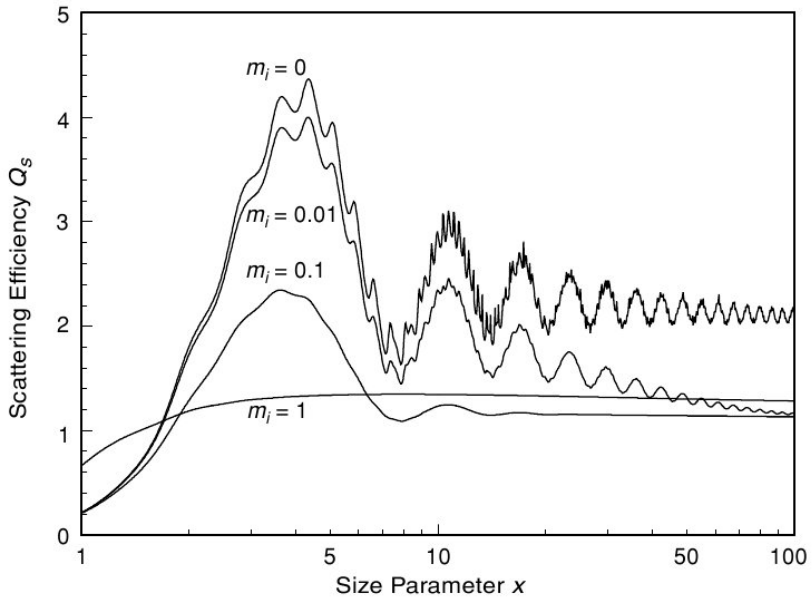


Figure 3: Efficiency factor for scattering Q_s as a function of the size parameter $x = 2\pi a/\lambda$. The real part of the refractive index used is $m_r = 1.5$, with results shown for 4 values of the imaginary part m_i.

- The relation between the incident and scattered Stokes parameters follows from Equation (36) [scatterer is a single sphere]:

$$\begin{bmatrix} I^{sca} \\ Q^{sca} \\ U^{sca} \\ V^{sca} \end{bmatrix} = \frac{1}{k^2 r^2} \begin{bmatrix} S_{11} & S_{12} & 0 & 0 \\ S_{12} & S_{11} & 0 & 0 \\ 0 & 0 & S_{33} & -S_{34} \\ 0 & 0 & S_{34} & S_{33} \end{bmatrix} \begin{bmatrix} I^{inc} \\ Q^{inc} \\ U^{inc} \\ V^{inc} \end{bmatrix} \tag{43}$$

$$S_{11} = \frac{1}{2}(|S_2|^2 + |S_2|^1) \qquad S_{12} = \frac{1}{2}(|S_2|^2 - |S_2|^1)$$
$$S_{33} = \frac{1}{2}(S_1 S_2^* + S_2 S_1^*) \qquad S_{12} = \frac{i}{2}(S_1 S_2^* - S_2 S_1^*) \tag{44}$$

- Only 3 of these 4 matrix elements are independent: $S_{11}^2 = S_{12}^2 + S_{33}^2 + S_{34}^2$

Scattering phase matrix elements

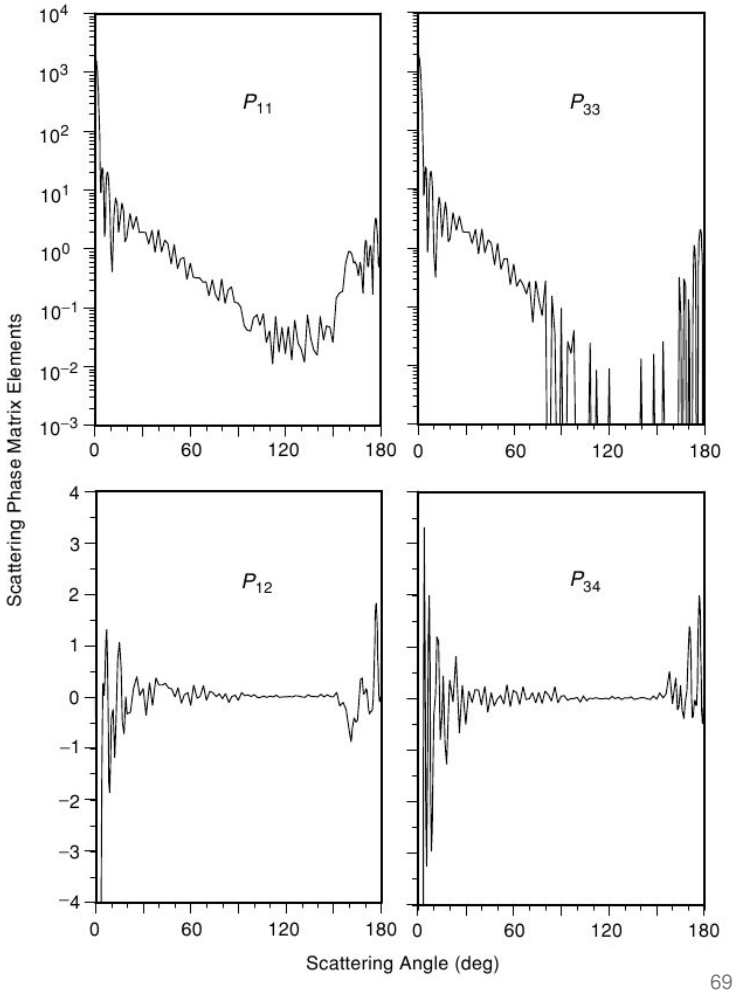


Figure 4: Scattering phase matrix elements as functions of the scattering angle for a refractive index $m_r = 1.5$ and a size parameter $x = 60$, computed from the Mie theory.