Scattering Angle

Scattering angle Θ depends on direction of incidence (θ_i,ϕ_i) and direction of scattering (θ_s,ϕ_s): $\cos\Theta = \cos\theta_i\cos\theta_s + \sin\theta_i\sin\theta_s \cos(\phi_s - \phi_i)$

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).

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$r$.

Moments: $$\int_0^\infty r^k n(r)dr = Nr_0^k \exp\left(k^2\sigma^2/2\right)$$

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.

Scattering

Matter is composed of discrete electric charges which are excited (set to oscillated) in presence of EM field.
Oscillating charges radiate EM waves, called *scattered* waves.
Incident waves form source excite secondary waves from scatterer → what is observed is the superposition of all waves.
If frequency of scattered waves is identical to source, *elastic* scattering.

What do *reflection, refraction, interference, diffraction, scattering* have in common?
All are scattering phenomena.

Scattering is the consequence of excitation of charges in ***matter***, not just at/near the surface.

Diffraction from slit: the slit (absence of matter/empty space) is not the source of the diffraction pattern.

Scattering by a dipole (The simplest example of an electrically neutral system.)
Equation of motion for a harmonic oscillator, mass m, charge e, acted on by EM wave with frequency ω: $m\frac{d^2\mathbf{x}}{dt^2} = -K\mathbf{x} - b\frac{d\mathbf{x}}{dt} + w\frac{d^3\mathbf{x}}{dt^3} + e\mathbf{E_o}(-i\omega t)$
K, *b*, and *w* are constants.

Terms on right side of equation:

1. Restoring force
2. Dissipative force
3. Radiative reaction
4. E-field force

Steady state solution: $\mathbf{x} = \mathbf{x}_o \exp(-i\omega t)$

where $\mathbf{x}_o = \frac{e}{m}\frac{\mathbf{E}_o}{\omega_o^2 - \omega^2 - i\gamma\omega}$, $\omega_o = \frac{K}{m}$, $\gamma = \gamma_a + \gamma_s\omega^2/\omega_o^2$, $\gamma_a = \frac{b}{m}$, and $\gamma_s = \omega_o\frac{w}{m}$

The ***instantaneous*** rate at which work is done on oscillator by external field: $P = e\mathbf{E}\cdot\frac{d\mathbf{x}}{dt}$

Time average: $\langle P\rangle = \langle P_a\rangle + \langle P_s\rangle$ = power absorbed plus power scattered

$\langle P_s\rangle = \frac{e^2}{2m}\frac{E_o^2}{\omega_o^2}\frac{\gamma_s\,\omega^4}{(\omega_o^2-\omega^2)^2+\gamma^2\omega^2}$ and $\langle P_a\rangle = \frac{e^2}{2m}E_o^2\frac{\gamma_a\,\omega^2}{(\omega_o^2-\omega^2)^2+\gamma^2\omega^2}$

Absorbed power: radiant energy transformed into other forms

Scattered power: incident radiant energy transformed into different directions → radiant energy is conserved.

Oscillation frequency of dipole and frequency of scattered radiation is equal to frequency of incident radiation: elastic scattering.

Note: scattering and absorption are not completely independent; both have dependence on $\gamma = \gamma_a + \gamma_s\omega^2/\omega_o^2$

Radiant power scattered in all directions $\langle P_s\rangle = \sigma_s F$

F is irradiance of beam that excites the scattered radiation.

Rayleigh scattering law: If $\omega \ll \omega_o$ and $\gamma \ll \omega_o$ then $\langle P_s\rangle \approx \frac{e^2}{2m}\frac{E_o^2}{\omega_o^6}\gamma_s^2\omega^4$

Note the λ^{-4} (ω^4) dependence

Rayleigh's Law

Perhaps the most famous result ever obtained by dimensional analysis:

The field E_s scattered by a particle small compared with wavelength of incident light is proportional to its volume V (number of dipoles or molecules) and to incident field E_i.

Energy conservation requires that the scattered field diminish inversely as the distance r from the particle; scattered power diminishes as r^2.

To make this proportionality dimensionally homogeneous requires the inverse square of a quantity with the dimensions of length. The only plausible physical variable at hand is the wavelength of the incident light, which leads to: $E_s \propto \frac{E_i V}{r\lambda^2}$

When the field is squared to obtain the scattered power, the result is Rayleigh's inverse fourth-power law.

Particle Scattering

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

To understand why requires understanding of interference → wave theory

Solution to the one-dimensional wave equation: $y = a \cos(kx - \omega t)$
wavenumber $k = 2\pi/\lambda$
$v = \omega/k$
$kx-\omega t$ is the phase; surfaces of constant phase are planes

Superposition and Interference
Wave equation is linear so sum (superposition) of solutions is a solution: if y_1 and y_2 are solutions, so is $a_1y_1 + a_2y_2$

Superposition of waves leads to *interference*
Plane waves with same frequency but different phase are superposed:

$\psi_2 = a_2 \exp(ikx - i\omega t + i\varphi_2)$ and $\psi_1 = a_1 \exp(ikx - i\omega t + i\varphi_1)$
Time averaged power: $\propto|\psi 1+ \psi 2|^2$
or $a_1^2 + a_2^2 + 2a_1a_2\cos(\varphi_2 - \varphi_1)$; last term is the interference term

$\cos(\Delta\varphi)$ lies between -1 and 1 so transmitted power lies between $(a_1 - a_2)^2$ and $(a_1 + a_2)^2$

- If $a_1 = a_2 = a$ then transmitted power is between 0 (destructive interference) and $4a^2$ (constructive interference).
- Interference is not *annihilation* nor *creation* of radiation. Radiation can only be annihilated (*absorbed*) or created (*emitted*) by matter.

Coherence

- Waves are coherent if they have fixed phase relationship – they *stick* together.
- Without fixed phase difference waves are incoherent: total power transmitted is sum of individual powers.
- Coherent waves arise from coherent sources: its elements have fixed spatial relationship.
- Coherence length (see section 3.4.2 on two slit interference): $\lambda r/2a$; *r* is distance to source, a is linear dimension
- If *d* represents linear dimension of an array of scatterers, array will give rise to interference pattern if $d << \lambda r/2a$.
- For visible light coherence length of sun is ~ 50 μm. 10 μm drops (<< coherence length) gives rise to the corona.

Phase shift on scattering

Coherence length of source >> distance between dipoles.

As distance to O increases indefinitely phase difference between scattered waves:

$$\Delta\varphi = \frac{2\pi}{\lambda}(\mathbf{e}_i - \mathbf{e}_s)\cdot\mathbf{r}_{12}$$

Forward direction is always in phase: $\Delta\varphi = 0$

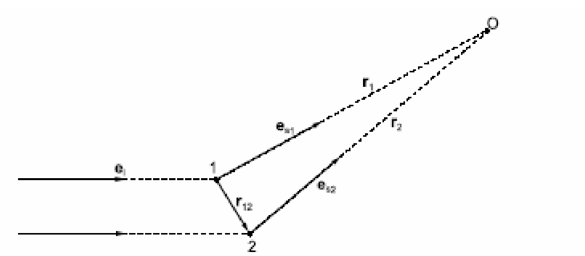


Figure 3.5: At O the total scattered field is the superposition of fields scattered by the two dipoles a fixed distance $\mathbf{r}_{12}$ apart. The phase difference between these two fields depends on the angle between the incident and scattered directions and the distance between the dipoles relative to the wavelength of the illumination.

Now consider a particle that is a coherent array of N dipoles and the linear dimension of this array is small compared to the wavelength. All scattered waved in any direction are in phase. Therefore, the power scattered in any direction is N^2 times the power scattered by one. (N ∝ *volume* → start of Rayleigh's dimensional analysis → so power proportional to *volume*^{2} for particles small compared to wavelength.)

- As size of particle increases phase differences between its dipoles increases
- Phase differences of order π become possible for some directions → in particular directions they may interfere destructively.
- Increase in scattering by v^2 with cannot be sustained indefinitely as particle grows.
- Consider two dipoles aligned with incident radiation from source:

$$\Delta\varphi = \frac{2\pi r}{\lambda}(1-\cos\Theta)$$, Θ is scattering angle

- $1-\cos\Theta$ goes from forward (0) to backward (2) direction.
- As r/λ increases so does number of oscillations in scattering as a function of angle

Scattering by Air and Liquid Water Molecules

$$|\psi|^2 = a^2 N + a^2 \sum_{j=1}^{N} \sum_{m\neq j}^{N} \exp(ik(\mathbf{e_i}-\mathbf{e_s})\mathbf{r}_{jm}$$

- N air molecules in a volume equal to the coherence length cubed.
- $|\Psi|^2$ is time-averaged power transmitted
- Air molecules move and their positions are almost completely uncorrelated: volume accessible to a molecule is about 1000 times its volume.
- all phase differences are equally probable such that the sum is approximately zero
- scattering by N (in a small volume) is N times scattering by one.
- This is true for all scattering directions except the forward direction for which scattering is in-phase regardless of the molecular separation relative to the wavelength.
- the ***in-phase*** scattering is the source of refraction

Frequency-Dependence of Scattering by Air Molecules

Color of skylight follows from Rayleigh's scattering law:

- Air molecules small compared with the wavelengths of visible light
- **another condition, the frequencies for which must be much less than a resonant frequency of the scatterer.**

What is the resonance frequency? Harmonic oscillator model: restoring force given by Coulomb's Law for attractive force between nucleus and electron.

$F = Kx = -\dfrac{q^2 x}{4\pi\varepsilon_o R^3}$ and $\omega_o = \sqrt{\dfrac{K}{m}} = \sqrt{\dfrac{q^2}{4\pi\varepsilon_o m R^3}}$, ε_o is the permittivity of free space,

m is the mass of the electron cloud. If we take R = 0.15nm for the atomic radius we obtain a resonant frequency that corresponds to a wavelength of about 0.1 μm, well into the ultraviolet.

- ω_o is a *plasma frequency*. At lower frequencies a medium is reflecting, at higher it is transmitting.

Particle Scattering Summary:

Rayleigh's Law (dipole scattering): $\langle P_s \rangle \approx \frac{e^2}{2m}\frac{E_o^2}{\omega_o^6}\gamma_s^2\omega^4$

Superposition, interference, and coherence: power $\propto a_1^2 + a_2^2 + 2a_1a_2\cos(\varphi_2 - \varphi_1)$

Phase shift on scattering from dipoles: $\Delta\varphi = \frac{2\pi}{\lambda}(\mathbf{e}_i - \mathbf{e}_s)\cdot\mathbf{r}_{12}$

As size of particle increases phase differences between its dipoles increases ($\mathbf{r}_{12}$ increases)
Phase differences of order π become possible for some directions → in particular directions they may interfere destructively
Increase in scattering by *volume*^{2} with cannot be sustained indefinitely as particle grows.
Consider two dipoles aligned with incident radiation from source: $\Delta\varphi = \frac{2\pi r}{\lambda}(1-\cos\Theta)$
1-cosΘ goes from forward (0) to backward (2) direction.
As r/λ increases so does number of oscillations in scattering as a function of angle

Scattering by Particles

Use different symbols for molecular and particle scattering cross sections to emphasize that molecules and particles are different.

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:
$\beta = NC_{\text{sca}}$

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

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:

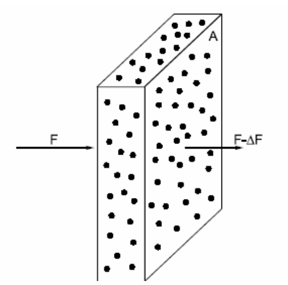
$Q_{sca} = \frac{C_{sca}}{G}$ $Q_{ext} = \frac{C_{ext}}{G}$

Scattering by Particles: slab approximation

How do scattering cross sections of particles depend on their ***size and the wavelength*** of the illumination?

- Consider slab of thickness h, area $A>>\lambda^2$, and refractive index n.
- The total power P_R reflected by this slab is F_0AR
- Coherent reflectivity: $\widetilde{R} = \dfrac{2R_\infty(1-\cos\varphi)}{1-2R_\infty\cos\varphi + R_\infty^2}$
- Phase difference between slab boundaries: $\varphi = \dfrac{4\pi nh}{\lambda}$
- Reflectivity of infinite slab is: $R_\infty = \left|\dfrac{n-1}{n+1}\right|^2$
- The reflection cross section is defined as P_R/F_0, and so reflection cross section per unit volume ($v = Ah$) for a sufficiently thin slab ($h<<\lambda$) is:

$$C_{ref}/\upsilon = \frac{\widetilde{R}}{h} \approx \frac{(n^2-1)^2\pi^2 h}{\lambda^2}$$



Scattering by Particles : slab approximation

- Increase thickness of slab by piling sheets of dipoles.
- Volumetric reflection cross section increases monotonically with each added sheet as long as all parts of the slab are excited in phase with all other parts.
- When total thickness reaches the point where appreciable (~π/2) phase differences occur the cross section decreases with increasing thickness
- Oscillations decrease with amplitude and occur with decreasing distance between adjacent peaks.

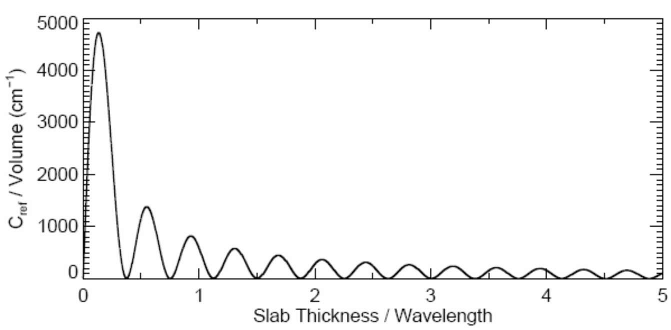


Figure 3.7: Reflection (scattering in the backward direction) cross section per unit volume of a transparent slab (with refractive index 1.33) illuminated at normal incidence as a function of slab thickness relative to the wavelength of the illumination (1 μm).

Complex Refractive Index

The theory of scattering and absorption by a homogeneous particle contains a single material parameter, its *complex refractive index.*
Why is it complex? Consider plane scalar wave propagating in +x direction:

$$\psi = a\exp(ikx - i\omega t)$$

Phase speed is ω/k and c is free-space speed of light: $\psi = a\exp\{i\omega(nx/c - t)\}$

where $n=c/v$, the refractive index of medium in which the wave propagates.

Wave does not attenuate if a = constant! Allow wave to attenuate exponentially by using complex amplitude $a\exp(-n''\omega x/c)$: $\psi = a\exp\{i\omega(Nx/c - t)\}$

where **N** *is* ***the complex refractive index***: $N = n' + in''$

Alternatively, can say that wavenumber is complex: $k = \frac{\omega}{c}N = k' + ik''$

Time averaged transmitted power: $\psi\psi^* = |\psi|^2 = a^2\exp\{-2n''\omega x/c\} = a^2\exp\{-4\pi n'' x/\lambda\}$

Now we have a relationship between absorption coefficient and imaginary refractive index: $\kappa = \frac{4\pi n''}{\lambda}$

Complex Refractive Indices of Liquid Water

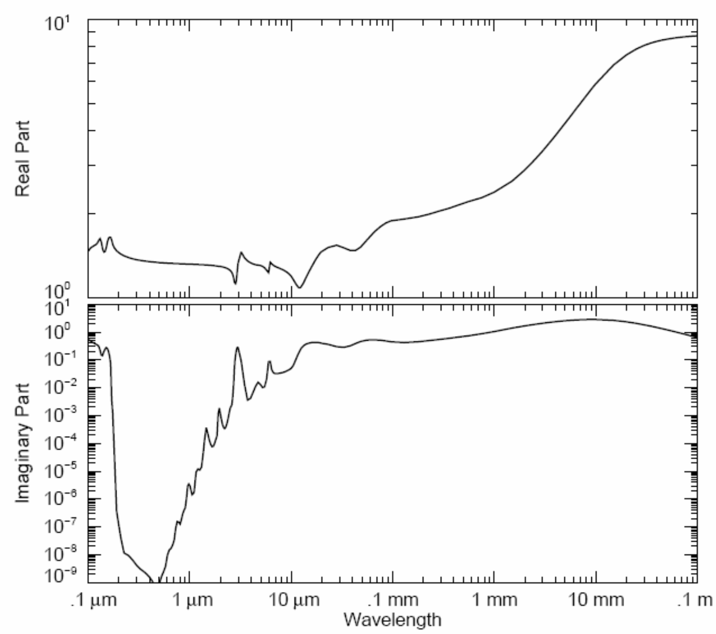


Figure 3.8: Optical constants (complex refractive index) of liquid water taken from the compilation by Querry et al. (1991) cited at the end of Chapter 2.

***Bohren and Clothiaux*, fig. 3.8**

Scattering by spheres

- Mie theory: scattering by arbitrary homogeneous sphere illuminated by a plane wave
- He was interested in explaining the color of colloidal gold
- Others preceded him, most notably Lorenz (not Lorentz)
- Mie scattering is a theory (one of many), not a physical process
- Not even exact because it is based on continuum EM theory and illumination by a plane wave of infinite lateral extent
- 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 → 0)
- Mie scattering by cylinders, spheroids, coated spheres? Mie never considered it; not "Mie theory."

Mie Theory

Scattering by arbitrary, homogeneous sphere illuminated by a plane wave:

- 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 chosen such that the tangential components of the fields are continuous across sphere surface
- Formally identical to problem of reflection and refraction but more complicated because scattered and internal fields are not plane waves
- Observable quantities are expressed in terms of the complex scattering coefficients a_n and b_n:

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

Scattering coefficients:

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

$\Psi_n(x)$ 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.
- Rule of thumb: number of terms required for convergence of series is ~ the size parameter ($2\pi r/\lambda$). 1 mm raindrops would require about 1000 terms

For x << 1 (Rayleigh limit):

$$C_{ext} = \frac{6\pi v}{\lambda}\Im\left\{\frac{m^2-1}{m^2+2}\left[1+\frac{4\pi^2 r^2}{15\lambda}\left(\frac{m^2-1}{m^2+2}\right)\frac{m^4+27m^2+38}{2m^2+3}\right]\right\}+\frac{24\pi^3 v^2}{\lambda^4}\left|\frac{m^2-1}{m^2+2}\right|^2$$

$$C_{sca} = \frac{24\pi^3 v^2}{\lambda^4}\left|\frac{m^2-1}{m^2+2}\right|^2 \qquad C_{abs} = \frac{6\pi v}{\lambda}\Im\left\{\frac{m^2-1}{m^2+2}\right\}$$

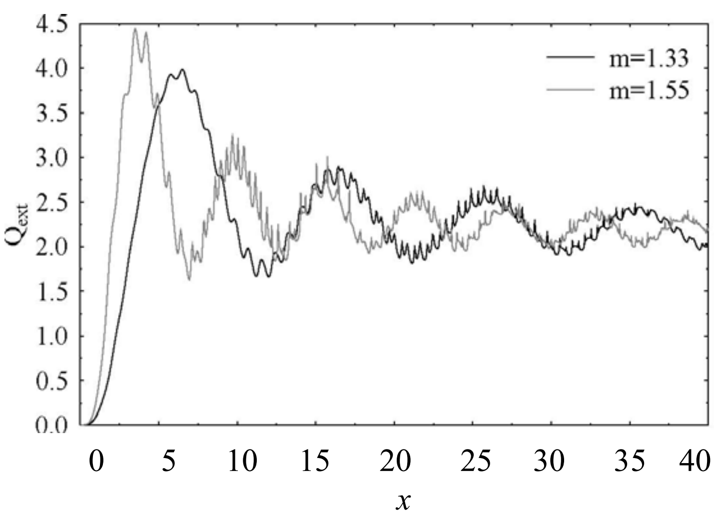
v is volume of sphere.

Mie Theory Results

Extinction efficiency versus size parameter (no absorption):

1) Small in small particle limit, Rayleigh's law: $Q_{ext} \propto x^4$.
2) Largest Q_{ext} when particle and wavelength have similar size.
3) $Q_{ext} \rightarrow 2$ in geometric limit ($x \rightarrow \infty$).
4) Oscillations from interference of transmitted and scattered waves:
   Phase difference $\Delta\phi$ between ray that traverses a large transparent sphere of radius r without deviation and the forward scattered light: $\Delta\phi = (2\pi/\lambda)2r(N_1\text{-}N)/N = 2x(m\text{-}1)$
   Condition for destructive interference:
   $\Delta\phi = (2p+1)\pi$, or $x(m\text{-}1)=(2p+1)\pi/2$; p=1,2, …
   Condition for constructive interference:
   $\Delta\phi = (2p+1)\pi/2$, or $x(m\text{-}1)=(2p+1)\pi/4$; p=1,2, …
5) Ripple structure from surface waves -resonance effects

Interference and ripple structure



Absorption damps interference and ripple structure: