6. LIGHT SCATTERING

6.1 The first Born approximation

- In many situations, light interacts with inhomogeneous systems, in which case the generic light-matter interaction process is referred to as *scattering*

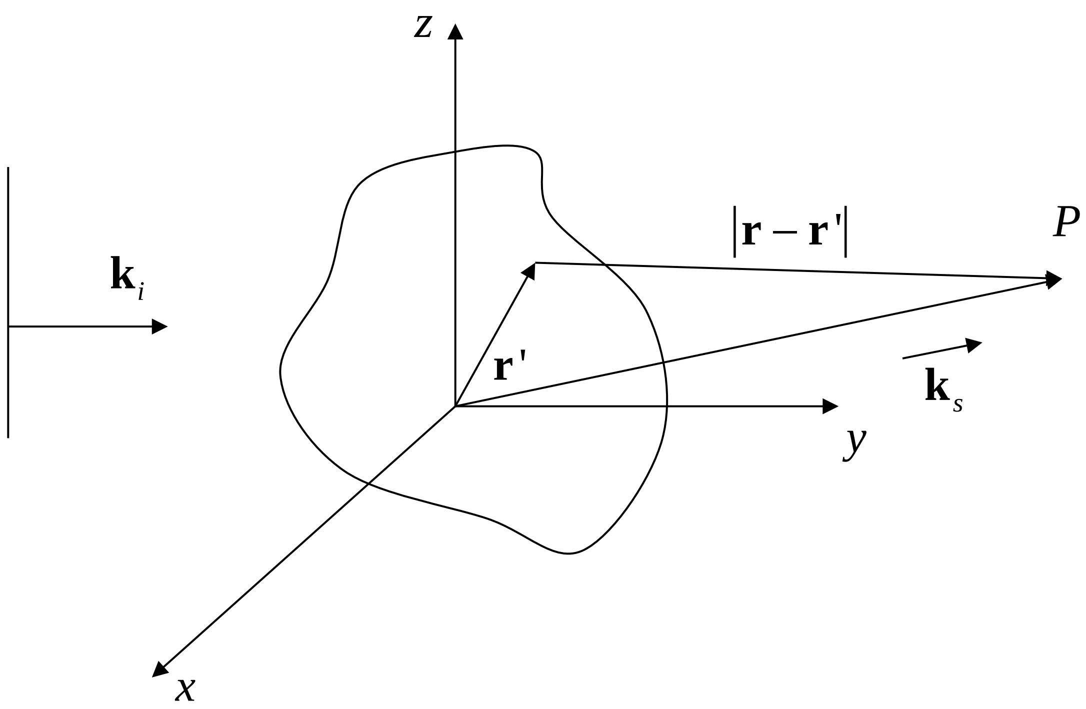

Figure 6-1. Light scattering by an inhomogeneous medium.

- The goal in light scattering experiments is to infer information about the refractive index distribution $n(\mathbf{r})$ from measurements on the scattered light

- We show that this problem can be solved if we assume weakly scattering media.

- Recall the Helmholtz equation for a loss-free medium

$$\nabla^2 U(\mathbf{r},\omega)+\beta^2(\mathbf{r},\omega)U(\mathbf{r},\omega)=0$$
$$\beta(\mathbf{r},\omega)=n(\mathbf{r},\omega)k_0 \qquad (6.1)$$
$$k_0=\omega/c$$

- Equation 1a can be re-written to show the source term on the right hand side, such that the scattered field satisfies

$$\nabla^2 U(\mathbf{r},\omega)+k^2{}_0 U(\mathbf{r},\omega)=-4\pi F(\mathbf{r},\omega)\cdot U(\mathbf{r},\omega)$$
$$F(\mathbf{r},\omega)=\frac{1}{4\pi}k^2{}_0\left[n^2(\mathbf{r},\omega)-1\right] \qquad (6.2)$$

- $F(\mathbf{r},\omega)$ is called the *scattering potential* associated with the medium.

- The elementary equation that gives Green's function $h(\mathbf{r},\omega)$ is

$$\nabla^2 h(\mathbf{r},\omega) + k^2{}_0 h(\mathbf{r},\omega) = -\delta^{(2)}(\mathbf{r}) \qquad (6.3)$$

- The solution in the spatial frequency domain is

$$\tilde{h}(\boldsymbol{k},\omega) = \frac{1}{k^2 - k^2{}_0} \qquad (6.4)$$

- Taking the Fourier transform of Eq. 4, we arrive at the spherical wave solution

$$\mathbf{h}(\mathbf{r},\omega) = \frac{e^{ik_0 r}}{r}. \qquad (6.5)$$

- The solution for the scattered field is a convolution between the source term and the Green function, $h(\mathbf{r},\omega)$,

$$U(\mathbf{r},\omega) = \int F(\mathbf{r}',\omega)\cdot U(\mathbf{r}',\omega)\cdot \frac{e^{ik_0|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|} d^3\mathbf{r}' \qquad (6.6)$$

- The integral in Eq. 6 can be simplified if we assume that the measurements are performed in the *far-zone*, $r' \ll r$. Thus, we can write

$$
\begin{aligned}
\left|\mathbf{r}-\mathbf{r}'\right| &= \sqrt{r^2 + r'^2 - 2\mathbf{r}\mathbf{r}'} \\
&\simeq r - \frac{\mathbf{r}}{r}\cdot\mathbf{r}' \qquad (6.7) \\
&\simeq r - \frac{\mathbf{k}_s}{k_0}\cdot\mathbf{r}'
\end{aligned}
$$

- $\mathbf{r}\cdot\mathbf{r}'$ is the scalar product of vectors $\mathbf{r}$ and $\mathbf{r}'$ and $\mathbf{k}_s / k_0$ is the unit vector associated with the direction of propagation.

- With this *far-zone* approximation, Eq. 6 can be re-written as

$$U(\mathbf{r},\omega) = \frac{e^{ik_0 r}}{r}\int F(\mathbf{r}',\omega)\cdot U(\mathbf{r}',\omega)\cdot e^{-i\cdot\mathbf{k}_s\cdot\mathbf{r}'} d^3\mathbf{r}' \qquad (6.8)$$

- Equation 8 indicates that, far from the scattering medium, the field behaves as a spherical wave, $\frac{e^{ik_0 r}}{r}$, perturbed by the *scattering amplitude*, defined as

$$f\left(\mathbf{k}_s,\omega\right)=\int F\left(\mathbf{r}',\omega\right)\cdot U\left(\mathbf{r}',\omega\right)\cdot e^{-i\mathbf{k}_s\cdot\mathbf{r}'}d^3\mathbf{r}' \qquad (6.9)$$

- To obtain a tractable expression for this integral, assume the scattering is *weak*, which allows us to expand $U\left(\mathbf{r}',\omega\right)$. The *Born approximation* assumes that the field inside the scattering volume is constant and equal to the incident field,

$$U_i\left(\mathbf{r}',\omega\right)\simeq e^{i\mathbf{k}_i\mathbf{r}'}. \qquad (6.10)$$

- With this approximation, we finally obtain for the scattered field

$$f\left(\mathbf{k}_s,\omega\right)=\int F\left(\mathbf{r}',\omega\right)\cdot e^{-i\left(\mathbf{k}_s-\mathbf{k}_i\right)\mathbf{r}'}d^3r' \qquad (6.11)$$

- The right hand side is a 3D Fourier transform.

- Within the first Born approximation, measurements of the field scattered at a given angle, gives access to the Fourier component $\mathbf{q} = \mathbf{k}_s - \mathbf{k}_i$ of the scattering potential *F*,

$$f(\mathbf{q}, \omega) = \int F(\mathbf{r}', \omega) \cdot e^{-i\mathbf{q}\mathbf{r}'} d^3\mathbf{r}'. \qquad (6.12)$$

- **q** is the difference between the scattered and initial wavevectors, called *scattering wavevector* and in quantum mechanics is referred to as the *momentum transfer*.

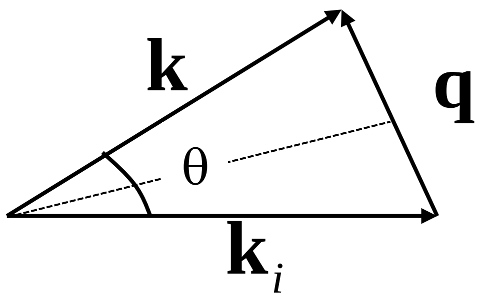

Figure 2. The momentum transfer.

- From Fig. 2 it can be seen that $q = 2k_0 \cdot \sin \theta/2$, θ the scattering angle. Due to the Fourier integral, Eq. 12 is easily invertible to provide the scattering potential,

$$F(\mathbf{r}',\omega) \propto \int U(\mathbf{q},\omega) \cdot e^{i\mathbf{q}\mathbf{r}'} d^3\mathbf{q}. \qquad (6.13)$$

- Equation 13 establishes the solution to the *inverse scattering problem*, it provides a way of retrieving information about the medium under investigation via angular scattering measurements. Measuring U at many scattering angles allows the reconstruction of F from its Fourier components.
- For *far-zone measurements* at a fixed distance R, the scattering amplitude and the scattered field differ only by a constant e^{ikR}/R, can be used interchangeably.

- To retrieve $F(\mathbf{r}',\omega)$ experimentally, two essential conditions must be met:
  - The measurement has to provide the complex scattered field
  - The scattered field has to be measured over an infinite range of spatial frequencies q (the limits of the integral in Eq. 13 are $-\infty$ to ∞)
- Finally, although good progress has been made recently in terms of measuring phase information, most measurements are intensity-based. Note that if one has access only to the intensity of the scattered light, $\left|U(q,\omega)\right|^2$, then the autocorrelation of $F(\mathbf{r}',\omega)$ is retrieved, and not F itself,

$$\int \left|U(q,\omega)\right|^2 \cdot e^{i\mathbf{qr}'} d^3\mathbf{q} = F(r',\omega) \otimes F(r',\omega) \quad (6.14)$$

- The result in Eq. 14 is simply the *correlation theorem* applied to 3D Fourier transforms.

Secondly, we have experimental access to limited frequency range, or bandwidth. The spatial frequency (or momentum transfer) range is intrinsically limited. Thus, for a given incident wave vector $\mathbf{k}_i$, with $k_i = k_0$, the highest possible $\mathbf{q}$ is obtained for backscattering, $\left|\mathbf{k}_b - \mathbf{k}_i\right| = 2k_0$ (Fig. 3a)

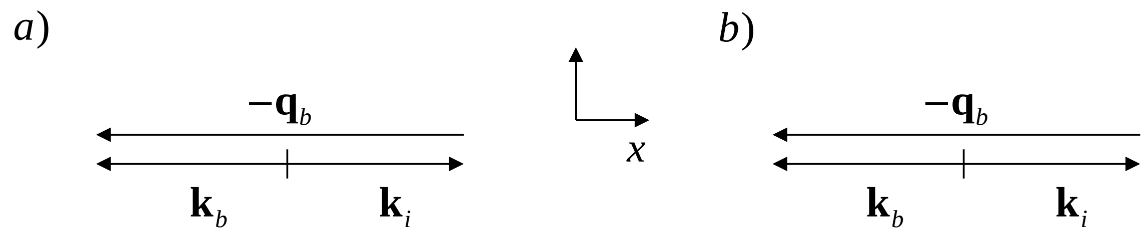

Figure 6-3. Momentum transfer for backscattering configuration for $\mathbf{k}_i \parallel \hat{x}$ *and* $\mathbf{k}_i \parallel -\hat{x}$.

Similarly for an incident wave vector in the opposite direction, $-\mathbf{k}_i$, the maximum momentum transfer is also $q = 2k_0$ (Fig. 3b). Altogether, the maximum frequency coverage is $4k_0$, as illustrated in Fig. 4.

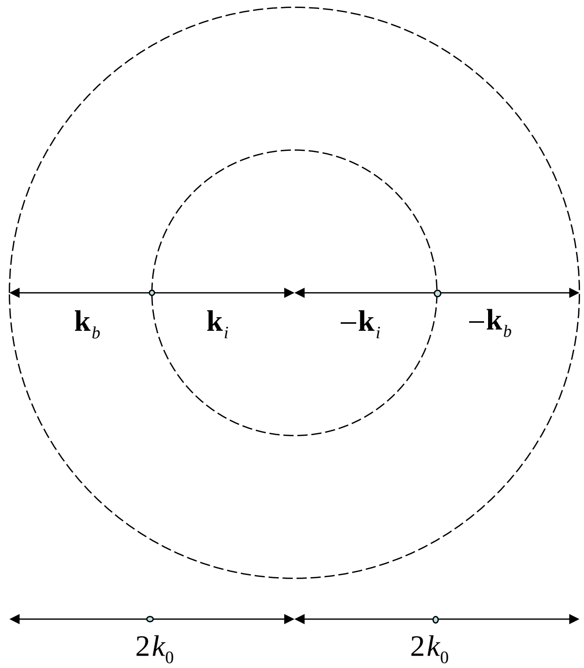

Figure 6-4. Ewald scattering sphere.

As we rotate the incident wavevector from $\mathbf{k}_i$ to $-\mathbf{k}_i$, the backscattering wavevector rotates from $\mathbf{k}_b$ to $-\mathbf{k}_b$, such that the tip of $\mathbf{q}$ describes a sphere of radius $2k_0$. This is known as the *Ewald sphere, or Ewald limiting sphere*.

Let us study the effect of this bandwidth limitation in the best case scenario of Ewald's sphere coverage. The measured (truncated field), $\underline{U}(\mathbf{q},\omega)$, can be expressed as

$$\underline{U}(\mathbf{q},\omega)=\begin{vmatrix} U(\mathbf{q},\omega), 0 \le q \le 2k_0 \\ 0, \text{ rest} \end{vmatrix} \tag{6.15}$$

Using the definition of a rectangular function in 3D, the "ball" function, we can express $\underline{U}$ as

$$\underline{U}(\mathbf{q},\omega)=U(\mathbf{q},\omega)\cdot\Pi\left(\frac{q}{4k_0}\right) \tag{6.16}$$

Thus, the scattering potential retrieved by measuring the scattered field $\underline{U}$ can be written as (from Eq. 13)

$$\underline{F}(\mathbf{r}',\omega)=F(\mathbf{r}',\omega)*\tilde{\Pi}(r') \tag{6.17}$$

In Eq. 17, $\tilde{\Pi}$ is the Fourier transform of the ball function $\Pi\left(\frac{q}{4k_0}\right)$, which has the form

$$\tilde{\Pi}(r') = \frac{\sin(2k_0 r') - 2k_0 r' \cos(2k_0 r')}{(2k_0 r)^3} \cdot (2k_0)^3 \quad (6.18)$$

It follows that even in the best case scenario, i.e. full Ewald sphere coverage, the reconstructed object $F(\mathbf{r}', \omega)$ is a "smooth" version of the original object, where the smoothing function is $\Pi(r')$. Practically, to cover the entire Ewald sphere, requires illuminating the object from all directions and measure the scattered complex field over the entire solid angle, for each illumination. This is, of course, a challenging task, rarely achieved in practice. Instead, fewer measurements are performed, at the expense of degrading resolution.

Figure 5 depicts the 1D profile of the 3D function $\Pi(r')$.

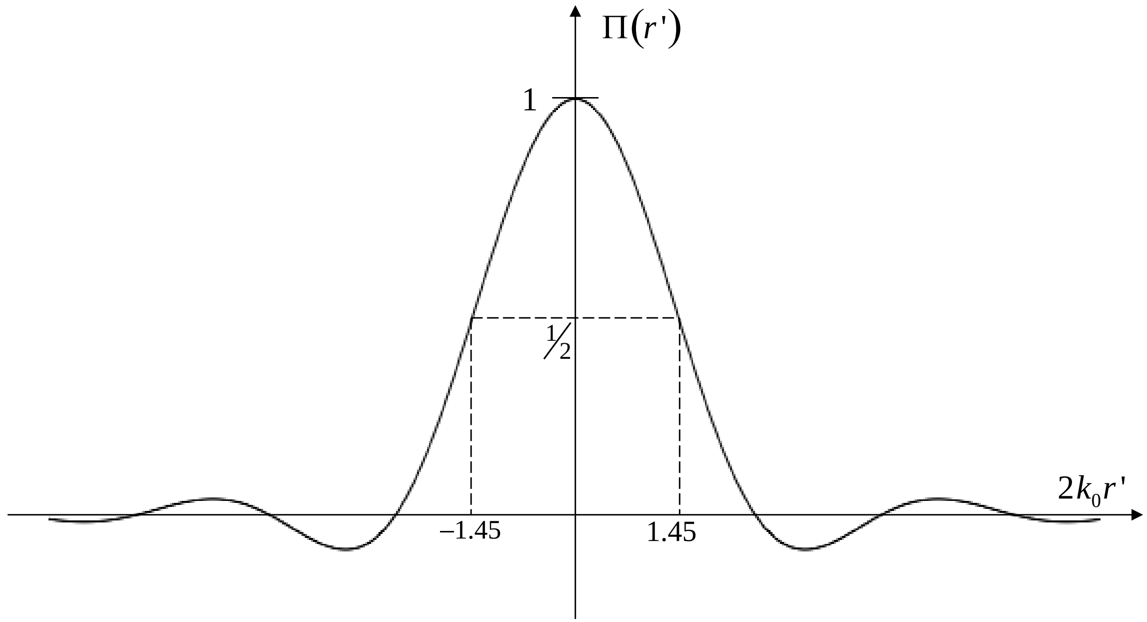

Figure 6-5. Profile through function $\Pi(r')$ *in Eq. 18.*

The FWHM of this function, $\Delta r'$, is given by $2k_0\Delta r' = 2q$, or $\Delta r' = 0.23\lambda$. We can, therefore, conclude that the best achievable resolution in reconstructing the 3D object is approximately $\lambda / 4$, only a factor of 2 below the common half-wavelength limit. This factor of 2 gain is dues to illumination from both sides.

6.2 Scattering cross sections of single particles.

Here by particle, we mean a region in space that is characterized by a dielectric permeability $\varepsilon = n^2 \in \mathbb{C}$, which is different from that of the surrounding medium (Fig. 6).

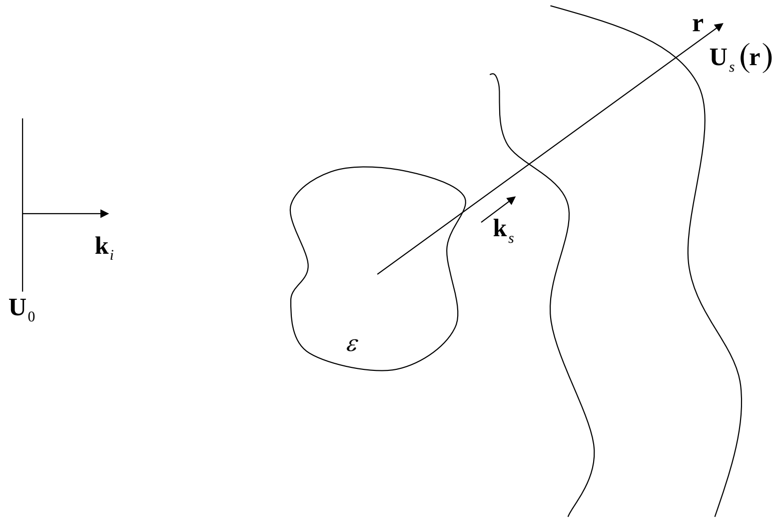

Figure 6-6. Light scattering by a single particle.

The field scattered in the far zone has the general form of a *perturbed* spherical wave,

$$\mathbf{U}_s(\mathbf{r}) = \mathbf{U}_0 \cdot \frac{e^{ikr}}{r} \cdot f(\mathbf{k}_s, \mathbf{k}_i), \tag{6.19}$$

where $\mathbf{U}_s(\mathbf{r})$ is the scattered field at position $\mathbf{r}$, $r = |\mathbf{r}|$, and $f(\mathbf{k}_s, \mathbf{k}_i)$ defines the *scattering amplitude*. The function f is physically similar to that encountered above, when discussing the Born approximation, (Eq. 9) except that here it also includes polarization in formation, in addition to the amplitude and phase.

The *differential cross-section* associated with the particle is defined as

$$\sigma_d(\mathbf{k}_s, \mathbf{k}_i) = \lim_{r \to \infty} r^2 \left| \frac{\mathbf{S}_s}{\mathbf{S}_i} \right|, \tag{6.20}$$

where $\mathbf{S}_s$ and $\mathbf{S}_i$ are the Poynting vectors associated along the scattered and initial direction, respectively, with moduli $|\mathbf{S}_{i,s}| = \frac{1}{2\eta}|U_{i,s}|^2$.

Note that, according to Eq. 20, σ_d is defined in the far-zone. It follows immediately that the differential cross-section equals the modulus squared of the scattering amplitude,

$$\sigma_d\left(\mathbf{k}_s, \mathbf{k}_i\right) = \left| f\left(\mathbf{k}_s, \mathbf{k}_i\right)\right|^2. \qquad (6.21)$$

Note that the unit for σ_d is $\left[\sigma_d\right] = m^2 / rad$.

One particular case is obtained for backscattering, when $\mathbf{k}_s = -\mathbf{k}_i$,

$$\sigma_b = \sigma_d\left(-\mathbf{k}_i, \mathbf{k}_i\right), \qquad (6.22)$$

where σ_b is referred to as the *backscattering cross section*.

The normalized version of σ_d defines the so-called *phase function*,

$$p\left(\mathbf{k}_s, \mathbf{k}_i\right) = 4\pi \frac{\sigma_d\left(\mathbf{k}_s, \mathbf{k}_i\right)}{\int_{4\pi} \sigma_d\left(\mathbf{k}_s, \mathbf{k}_i\right) d\Omega}. \qquad (6.23)$$

The phase function p defines the angular probability density associated with the scattered light. The integral in the denominator of Eq. 22 defines the *scattering cross section*.

$$\sigma_s = \int_{4\pi} \sigma_d \left(\mathbf{k}_s, \mathbf{k}_i\right) d\Omega \qquad (6.24)$$

Thus, the unit of the scattering cross section is $\left[\sigma_s\right] = m^2$. In general, if the particle also absorbs light, we can define an analogous *absorption cross section*, such that the attenuation due to the combined effect is governed by a *total cross section*,

$$\sigma = \sigma_a + \sigma_s \qquad (6.25)$$

For particles of arbitrary shapes, sizes, and refractive indices, deriving expression for the scattering cross sections, σ_d and σ_s, is very difficult. However, if simplifying assumptions can be made, the problem becomes tractable. In the following, we will investigate some of these particular cases, most commonly encountered in practice.

6.3 Particles under the Born approximation

When the refractive index of a particle is only slightly different from that of the surrounding medium, its scattering properties can be derived analytically within the framework of the Born approximation. Thus the scattered amplitude from such a particle is the Fourier transform of the scattering potential of the particle (Eq. 12)

$$f(\mathbf{q},\omega) = \int F_p(\mathbf{r}')\cdot e^{i\mathbf{q}\mathbf{r}'} d^3\mathbf{r}', \qquad (6.26)$$

In Eq. 26, F_p is the scattering potential of the particle. We can conclude that the scattering amplitude and the scattering potential form a Fourier pair, up to a constant k_0

$$f(\mathbf{q},\omega) \rightarrow \frac{1}{k_0}\cdot F(\mathbf{r}',\omega) \qquad (6.27)$$

The rule of thumb for this scattering regime to apply is that the total phase shift accumulation through the particle is small, say, $<1\,rad$. For a particle of diameter d and refractive index n in air, this condition is $(n-1)k_0 d<1$. Under these conditions, the problem becomes easily tractable for arbitrarily shaped particles. For intricate shapes, the 3D Fourier transform in Eq. 26 can be at least solved numerically. For some regular shapes, we can find the scattered field in analytic form, as described below.

6.3.1 Spherical particles

For a spherical particle, the scattering potential has the form of the *ball* function

$$F_p(\mathbf{r}')=\Pi\left(\frac{r'}{2r}\right)\cdot F_0$$
$$F_0=\frac{1}{4\pi}k_0{}^2\left(n^2-1\right) \qquad (6.28)$$

Equations 28a-b establishes that the particle is spherical in shape and is characterized by a constant scattering potential F_0 inside (and zero outside)

Thus, plugging Eqs. 28 into Eq. 26, we obtain the scattering amplitude distribution,

$$f(\mathbf{q},\omega) \propto (n^2-1) k_0{}^2 \cdot r^3 \cdot \frac{\sin(qr) - qr \cdot \cos(qr)}{(qr)^3} \quad (6.29)$$

The differential cross section is

$$\begin{aligned} \sigma_d(\mathbf{q},\omega) &= \left| f(\mathbf{q},\omega) \right|^2 \\ &\propto (n^2-1)^2 V^2 k_0{}^4 \left[\frac{\sin(qr) - qr \cdot \cos(qr)}{(qr)^3} \right]^2 \end{aligned} \quad (6.30)$$

where $V = 4\pi r^3/3$ is the volume of the particle.

Figure 7 illustrates the angular scattering according to Eq. 28.

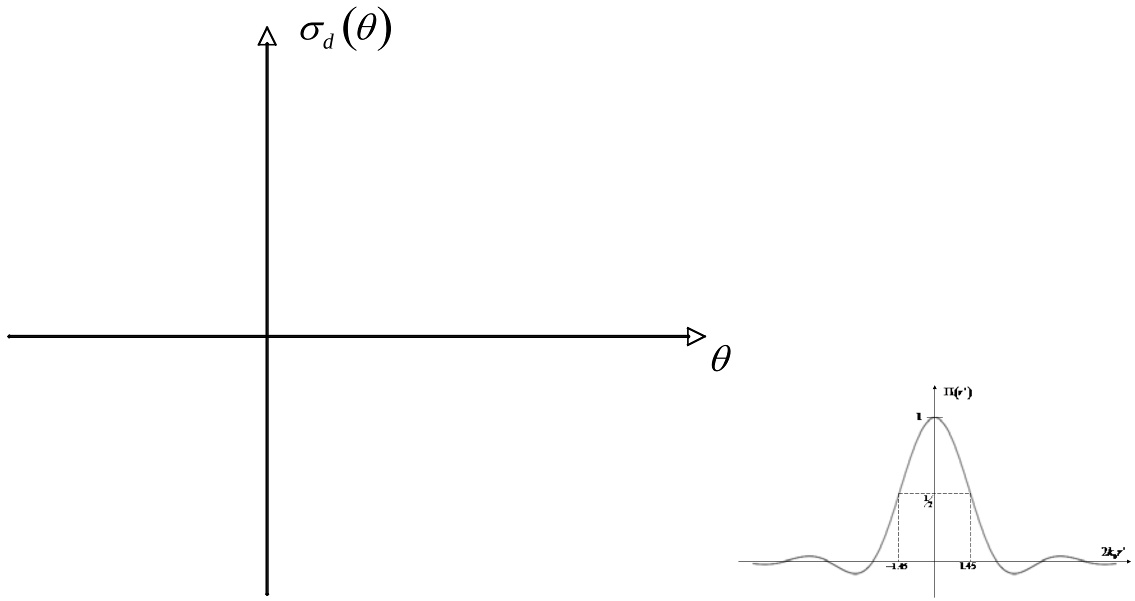

Figure 6-7. Angular scattering for a Rayleigh-Gauss particle with $qr = \sin \theta/2$.

Equation 30 establishes the differential cross section associated with a spherical particle under the Born approximation. Sometimes this scattering regime is referred to as Rayleigh-Gauss, and the particle for which this formula holds as Rayleigh-

Gauss particles. The scattering angle θ enters explicitly Eq. 28, by expressing $q = 2k_0 \sin \theta/2$.

A very interesting particular case is obtained when $qr \to 0$. Note that this asymptotical case may happen when the particle is very small, but also when the measurement is performed at very small angles, i.e. $\theta \to 0$. If we expand around the origin the function,

$$
\begin{aligned}
f(x) &= \left(\frac{\sin x - x \cdot \cos x}{x^3} \right)^2 \\
&\simeq \left(\frac{x - \frac{x^3}{6} - x\left(1 - \frac{x^2}{2}\right)}{x^3} \right)^2 \qquad (6.31) \\
&= \frac{1}{6}
\end{aligned}
$$

Thus, measurements at small scattering angles, can reveal volume of the particle,

$$\sigma_d(\mathbf{q},\omega)\Big|_{q\to 0} \propto \left(n^2-1\right)^2 V^2 k_0^{\ 4} \qquad (6.32)$$

This result is the basis for many flow cytometry instruments, where the volume of cells is estimated via measurements of *forward scattering*. The cell structure, i.e. higher frequency components are retrieved through larger angle, or *side scattering*.

On the other hand, Eq. 31 applies equally well where the particle is very small, i.e. Rayleigh particle. In this case, σ_d is independent of angles, i.e. Rayleigh scattering is isotropic. Thus, the scattering cross section for Rayleigh particles has the form

$$\sigma_s(\omega) \propto (n-1)^2 k_0^{\ 4} V^2. \qquad (6.33)$$

The scattering cross section of Rayleigh particles is characterized by strong dependence on size and wavelength

$$\sigma_s \propto r^6$$
$$\sigma_s \propto \lambda^{-4} \qquad (6.34)$$

The nanoparticles in the atmosphere have scattering cross sections that are 16 times larger for a wavelength $\lambda_b = 400nm$ (blue) than for $\lambda_r = 800nm$ (red). This explains why the clear sky looks bluish, and the sun itself looks reddish.

6.3.2 Cubical particles

For a cubical particle, the scattering potential has the form

$$F\left(x,y,z\right) = \Pi\left(\frac{x}{2a}\right)\cdot\Pi\left(\frac{y}{2b}\right)\cdot\Pi\left(\frac{z}{2c}\right)\cdot F_0$$
$$F_0 = \frac{1}{4\pi}k_0^{\ 2}\left(n^2-1\right) \qquad (6.35)$$

The scattering amplitude in this case is

$$f\left(q_x,q_y,q_z\right) = \left(n^2-1\right)k_0^{\ 2}V\,\mathrm{sinc}\left(q_x a\right)\cdot\mathrm{sinc}\left(q_y b\right)\cdot\mathrm{sinc}\left(q_z c\right) \qquad (6.36)$$

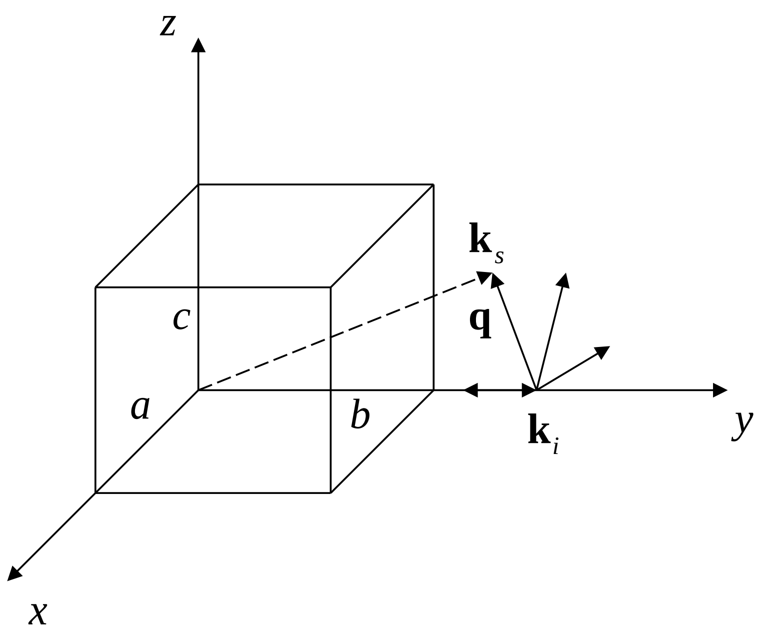

Figure 6-8. Scattering by a cubical particle.

It can be seen that the differential cross section $\sigma_d = |f|^2$ has the same V^4 and $k_0{}^4$ dependence as for spherical particles.

6.3.3 Cylindrical particles

For a cylindrical particle, the scattering potential is

$$F\left(x,y,z\right)=\Pi\left(\frac{\sqrt{x^2+y^2}}{2a}\right)\cdot\Pi\left(\frac{z}{2b}\right)\cdot F_0 \qquad (6.37)$$

$$F_0=\frac{k_0{}^2}{4\pi}\left(k^2-1\right)$$

The 3D Fourier transform of F yields the scattering amplitude

$$f\left(q_x,q_y,q_z\right)=\pi a^2 b\cdot\frac{J_1\left(\sqrt{q_x{}^2+q_y{}^2}\cdot a\right)}{2\sqrt{q_x{}^2+q_y{}^2}}\cdot \operatorname{sinc} q_z\cdot b \qquad (6.38)$$

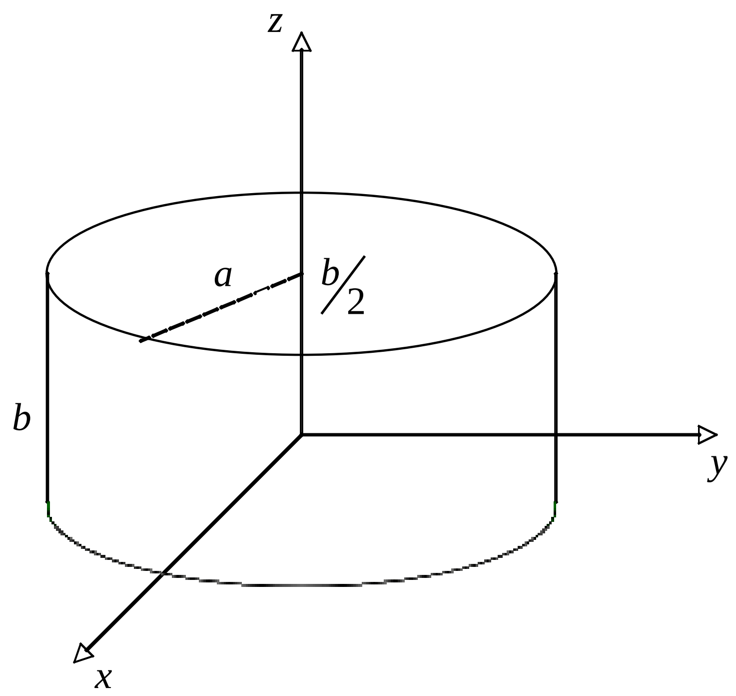

Figure 6-9. Scattering by a cylindrical particle.

As before, σ_d and σ_s can be easily obtained from $|f|^2$.

6.4 Scattering from ensembles of particles under the Born approximation.

Let us consider the situation where the scattering experiment is performed over an ensemble of particles randomly distributed in space, as illustrated in Fig. 10.

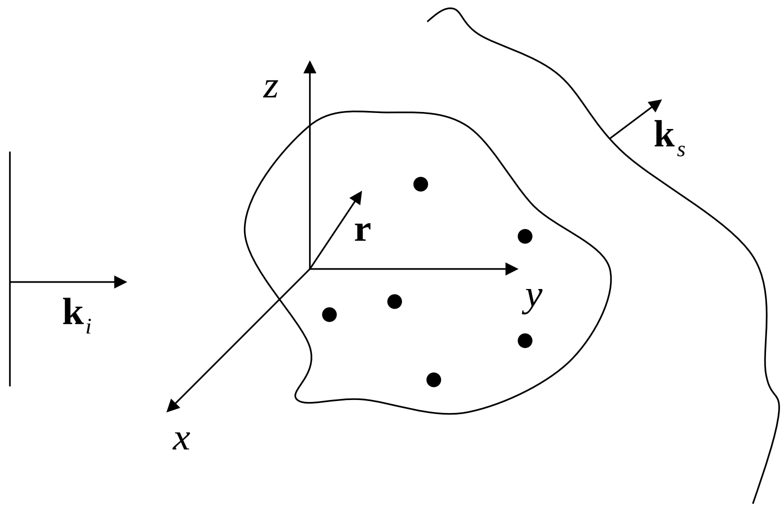

Figure 6-10. Scattering by an ensemble of particles.

If we assume that the ensemble is made of identical particles of scattering potential $F_0(\mathbf{r})$, then the ensemble scattering potential can be expressed via δ-functions as

$$F(\mathbf{r}) = F_0(\mathbf{r}) * \sum_j \delta(\mathbf{r} - \mathbf{r}_j) \tag{6.39}$$

Equation 39 establishes the discrete distribution of the scattering potential, where each particle is positioned at position r_i. Note that, in order for the Born approximation to apply, the particle distribution is space, which eliminates the possibility of "overlapping" particles. The scattering amplitude is simply the 3D Fourier transform of $F(\mathbf{r})$,

$$\begin{aligned} f(\mathbf{q}) &= \int F_0(\mathbf{r}) * \sum \delta(\mathbf{r} - \mathbf{r}_j) e^{i\mathbf{qr}} d^3\mathbf{r} \\ &= f_0(\mathbf{q}) \cdot \sum e^{i\mathbf{qr}_j}, \end{aligned} \tag{6.40}$$

where, as before, $\mathbf{q} = \mathbf{k}_s - \mathbf{k}_i$.

Therefore, the scattering amplitude of the ensemble is the scattering amplitude of a single particle, $f_0(\mathbf{q})$, multiplied by the so-called *structure function*, defined as

$$S(q) = \sum_j e^{i\mathbf{q}\mathbf{k}_j} \qquad (6.41)$$

We can express the scattering amplitude as a product,

$$f(\mathbf{q}) = f_0(\mathbf{q}) \cdot S(q) \qquad (6.42)$$

In Eq. 42, $f_0(\mathbf{q})$ is sometimes referred to as the *form function*. The physical meaning of the form and structure functions becomes apparent if we note that the size of the particle is smaller than the inter-particle distance. It follows that $f_0(q)$ is a broader function than $S(q)$, or that $f_0^{(5)}$ acts as the envelope (*form*) of $f(\mathbf{q})$ and $S(q)$ as its rapidly varying component (*structure*).

Example Scattering from 2 spherical particles (radius a) separated by a distance (b) (Fig. 11).

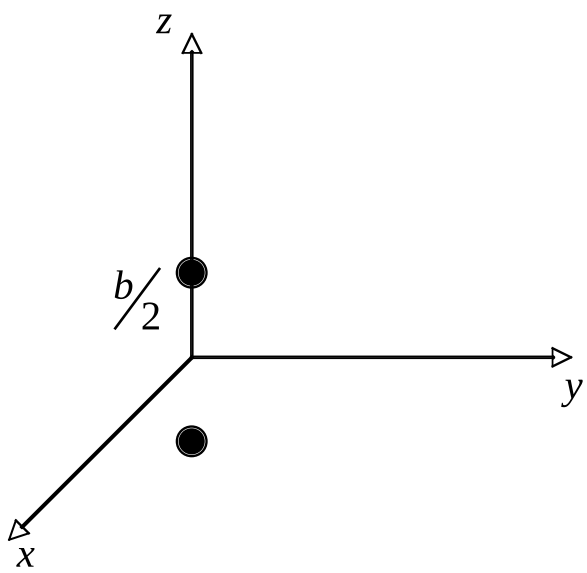

Figure 6-11. Scattering by two particles separated by a distance b.

According to Eq. 42, the far-zone scattering amplitude is

$$\begin{aligned} f(\mathbf{q}) &= f_0(\mathbf{q}) \cdot \cos\left(q_z \cdot b/2\right) \\ &= \left(n^2 - 1\right) k_0{}^2 V \cdot \frac{\sin(qr) - qr \cdot \cos(qr)}{(qr)^3} \cdot \cos\left(q_z \cdot b/2\right) \end{aligned} \qquad (6.43)$$

In Eq. 43 we wrote explicitly the form factor of a Rayleigh-Gauss particle of volume V and refractive index n.

This approach is the basis for extracting crystal structures form x-ray scattering measurements.

6.5. Mie scattering.

Mie provided the full electromagnetic solutions of Maxwell's equations for a *spherical particle* of arbitrary size and refractive index. The scattering cross section has the form

$$\sigma_s = \pi a^2 \frac{2}{\alpha^2} \sum_{n=1}^{\infty} (2n+1)\left(|a_n|^2 + |b_n|^2\right), \qquad (6.44)$$

where a_n and b_n are complicated functions of $k = k_0 a$, $\beta = k_0 n a / n_0$, with a the radius of the particle, k_0 the wavenumber in the medium, n_0 the refractive index of the medium, and n the refractive index of the particle. Equation 44 shows that the Mie solution is in terms of an infinite series, which can only be evaluated numerically. Although today common personal computers can evaluate σ_s very fast, we should note that as the particle increases in size, the summation converges more slowly, as a higher number of terms contribute significantly. Physically, as *a* increases, standing waves (nodes) with higher number of maxima and minima “fit” inside the sphere.

Although restricted to spherical particles, Mie theory is heavily used for tissue modeling.

6.6. Multiple Scattering Regime

- In the previous section, we introduced the basic description of the single scattering process, which applies to sparse distributions of particles. For the single scattering regime, the average dimension of the scattering medium must be smaller than the mean free path l_s.
- In many situations this is not the case and the light travels through the medium over distances much longer than l_s and encounters many scattering events during the propagation. For this case, the simple treatment presented earlier does not apply and the complexity of the problem is increased.
- A full vector solution to the field propagation in dense media is not available; scalar theories give results in an analytical form only for special, intermediate cases of multiple scattering regimes.

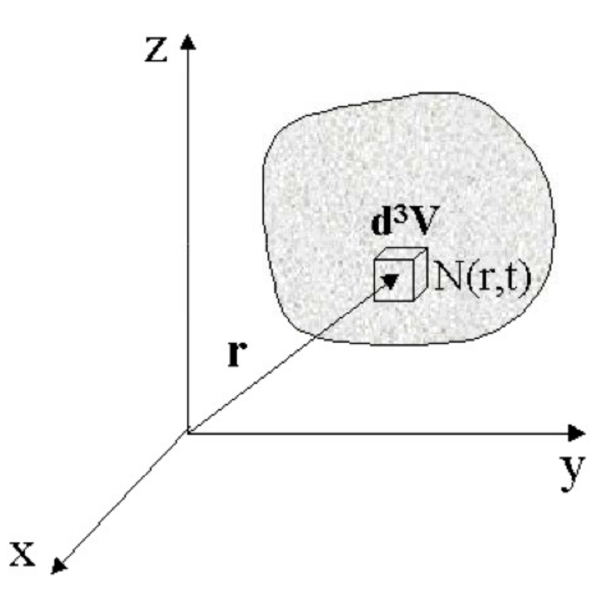

Figure 12. Scattering medium under transport conditions.

- Simplifications can be used when the scattering medium is very dense and the electromagnetic waves can be assumed to travel randomly through the medium.

6.6.1. Elements of Radiative Transport Theory

- The transport description in the case of a highly scattering medium is based on a photon random walk picture similar to that used in solid state physics and nuclear reactor physics [7]. The wave correlations that may survive this heavy scattering regime are neglected, while each scattering event is considered an independent process.
- Using the energy (photon) conservation principle, the final goal of the transport theory is to find the time-dependent photon distribution at each point in the scattering medium. We will make use of the following definitions and notations:
  - $N(r,t)$− photon density [m−3];
  - $v \cdot \mu_s = v/l_s$− frequency of interaction [s^{-1}];
  - $\mu = \mu_a + \mu_s$− attenuation coefficient [m];
  - $\Phi(r, t) = vN(\mathbf{r}, t)$− photon flux [$m^{-2} \cdot s^{-1}$];
  - $n(\mathbf{r},\mathbf{\Omega},t)$− density of photons ($m^{-3}sr^{-1}$) having the direction of propagation

within the solid angle interval [Ω, Ω + dΩ];

- φ(**r**, **Ω**, t)R= v · n(r, **Ω**, t)− angular photon flux [$m^{-2}s^{-1}sr^{-1}$]; scalar
- j(**r**, **Ω**, t) = **Ω** · φ(**r**, **Ω**, t)− angular current density [$m^{-2}s^{-1}sr^{-1}$]; vector
- J(**r**, t) = $\int_{4\pi} j(\mathbf{r},t,\Omega)d\Omega$ − photon current density [$m^{-2}s^{-1}$].

- To write the transport equation, we first identify all the contributions that affect the photon balance within a specific volume V. We obtain the following transport equation in its integral form:

$$\int_V \left\{ \begin{array}{l} \frac{dn}{dt} + v\Omega\nabla n + v\mu n(\mathbf{r},\mathbf{\Omega},t) - \int_{4\pi}\left[v'\mu_s(\Omega'\to\Omega)n(\mathbf{r},\mathbf{\Omega}',t)\right]d\Omega' - \\ -s(\mathbf{r},\mathbf{\Omega},t) \end{array} \right\} d^3r d\Omega = 0.$$

(6.45)

- The first term denotes the rate of change in the energy density, the second term represents the total leakage out of the boundary, the third term is the loss due to scattering, the fourth term accounts for the photons that are scattered into the direction $\mathbf{\Omega}$, while the last term represents the possible source of photons inside the volume.

- Since the volume V was arbitrarily chosen, the integral of Eq. 2.31 is zero if and only if the integrand vanishes. Thus, the differential form of the transport equation is:

$$\frac{dn}{dt} + v\Omega\nabla n + v\mu n(\mathbf{r},\mathbf{\Omega},t) - \\ -\int_{4\pi}\left[v'\mu_s(\Omega'\rightarrow\Omega)n(r,\Omega',t)\right]d\Omega' - s(\mathbf{r},\mathbf{\Omega},t) = 0. \quad (6.46)$$

- The same equation can be expressed in terms of the angular flux $\varphi(\mathbf{r}, \mathbf{\Omega}, t) = v \cdot n(\mathbf{r}, \mathbf{\Omega}, t)$:

$$\frac{1}{v}\frac{d\phi}{dt} + \Omega\nabla\phi + \mu\phi(\mathbf{r},\mathbf{\Omega},t) - \\ -\int_{4\pi}\left[\mu_s(\mathbf{\Omega}'\to\mathbf{\Omega})\phi(\mathbf{r},\mathbf{\Omega}',t)\right]d\mathbf{\Omega}' - s(\mathbf{r},\mathbf{\Omega},t) = 0. \quad (6.47)$$

- The initial condition for this equation is $\varphi(\mathbf{r}, \mathbf{\Omega}, 0) = \varphi_0(\mathbf{r}, \mathbf{\Omega})$, while the boundary condition is $\varphi(\mathbf{r}_s,\mathbf{\Omega},t) r_s \in S = 0$, if $\mathbf{\Omega} e_s < 0$. The transport equation can be solved in most cases only numerically. As we will see in the next section, further simplifications to this equation are capable of providing solutions in closed forms and will, therefore, receive a special attention.

6.6.2. The Diffusion Approximation of the Transport Theory

- Eq. 2.33 can be integrated with respect to the solid angle **Ω**, the result can be expressed in terms of the photon flux Φ and the current density J as a continuity equation:

$$\frac{1}{v}\frac{d\Phi}{dt} + \nabla \mathbf{J}(\mathbf{r},t) + \mu\Phi(\mathbf{r},t) = \mu_s\Phi(\mathbf{r},t) + s(\mathbf{r},t) \quad (6.48)$$

- Eq. 2.34 contains two independent variables Φ and J:

$$\Phi(\mathbf{r},t) = \int_{4\pi} \phi(\mathbf{r},\mathbf{\Omega},t)\,d\Omega$$
$$J(\mathbf{r},t) = \int_{4\pi} \mathbf{\Omega}\phi(\mathbf{r},\mathbf{\Omega},t)\,d\Omega \quad (6.49)$$

- An additional assumption is needed to make the computations tractable. In most cases, one can assume that the angular flux φ is only linearly anisotropic:

$$\phi(\mathbf{r},\mathbf{\Omega},t) \simeq \frac{1}{4\pi}\Phi(\mathbf{r},t) + \frac{3}{4\pi}J(\mathbf{r},t)\cdot\mathbf{\Omega}. \quad (6.50)$$

- This represents the first order Legendre expansion of the angular flux and is the approximation that lets us obtain the diffusion equation. It is called the P_1 approximation, since it represents a Legendre polynomial P_l, for l = 1.
- The photon source is considered to be isotropic and the photon current to vary slowly with time: $\frac{1}{|\mathbf{J}|}\frac{\partial \mathbf{J}}{\partial t} << v\mu_t$.
- The equation that relates J and Φ can be derived, which is known as Fick's law:

$$J(\mathbf{r},t) = -\frac{1}{3\mu_t}\cdot\nabla\Phi(\mathbf{r},t) = -D(r)\nabla\Phi(\mathbf{r},t) \quad (6.51)$$

- $\mu_t = \mu - g\mu_s$ is the transport coefficient. *g* represents the anisotropy factor and is defined in terms of elementary solid angles before and after scattering as:

$$g = \langle \mathbf{\Omega}\cdot\mathbf{\Omega}' \rangle. \quad (6.52)$$

- The anisotropy factor is the average cosine of the scattering angle, with the phase function defined in Eq. 2.6 as the weighting function. g accounts for the fact that, for large particles, the scattering pattern is not isotropic.

- In Eq. 2.37, D(r) is the diffusion coefficient of photons in the medium and is the only parameter that contains information about the scattering medium.
- Combining Eqs. 2.34 and 2.37, we can obtain the so-called diffusion equation

$$\frac{1}{v}\frac{\partial \Phi}{\partial t} - \nabla\left[D(r)\nabla\Phi(\mathbf{r},t)\right] + \mu_a(\mathbf{r})\Phi(\mathbf{r},t) = S(\mathbf{r},t).(6.53)$$

- μ_a is the absorption coefficient and S(**r**,t) is the photon source considered to be isotropic.
- The diffusion equation breaks down for short times of evolution, $t < l_t/c$. It takes a finite amount of time for the diffusion regime to establish.
- Although restricted to a particular situation of multiple light scattering, the diffusion treatment finds important applications in practice.

- Using appropriate boundary conditions, Eq. 2.39 can be solved for particular geometries. Once the photon flux Φ is calculated, the current density J can be obtained using Fick's law [7].
- The current density J(r,t) is the measurable quantity in a scattering experiment and thus deserves special attention. The total optical power measured by a detector is proportional with the integral of J(r,t) over the area of detection. *t* represents the time of flight for a photon between the moment of emission and that of detection. When a constant group velocity is assumed, this variable can be transformed into an equivalent one- the optical path-length, s = vt.
- The path-length probability density $P(s) = J(s) / \int_0^\infty J(s)ds$, the probability that the photons traveled, within the sample an equivalent optical path-length in the interval (s, s + ds), can also be evaluated.