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Reduced light-scattering properties for mixtures of spherical particles: a simple approximation derived from Mie calculations

R. Graaff, J. G. Aarnoudse, J. R. Zijp, P. M. A. Sloot, F. F. M. de Mul, J. Greve, and M. H. Koelink

The reduced scattering cross section per unit of volume \( \Sigma' \equiv \Sigma_s(1 - g) \) is an important parameter to describe light propagation in media with scattering and absorption. Mie calculations of the asymmetry factor \( g \) for nonabsorbing spheres and \( Q_s = \frac{\Sigma_s}{\Sigma} \), the ratio of the scattering cross section \( \Sigma_s \) and the particle cross section, show that
\[
Q_s(1 - g) = 3.28x^{0.37}(m - 1)^{2.09}
\]
is true to within a few percent, when the Mie parameters for relative refractive index \( m \) and size \( x \) are in the ranges of \( 1 < m < 1.1 \) and \( 5 < x < 50 \), respectively. A ratio of reduced scattering cross sections for radiation at two wavelengths is also independent of the size within the range mentioned, even for mixtures of different size spheres. The results seem promising for biomedical applications.

Key words: Mie scattering, multiple scattering, medicine, biology, light propagation in blood and tissue.

Introduction

Simulations of light scattering in media with scattering and partly absorbing particles such as spheres, and also such as red blood cells and scatterers in living tissues, can be performed by using Monte Carlo simulations or the transport theory. The basic properties of the scatterers, such as \( \sigma_a \) and \( \sigma_s \), the absorption and scattering cross sections, and \( g \), the asymmetry factor, which is the average cosine of the scattering angle, must be available. A reduced scattering cross section \( \sigma_s' = \sigma_s(1 - g) \) for a single scatterer can be defined.

In the diffusion approximation of transport theory, light propagation within a scattering medium with a given geometry can be fully described by \( \Sigma_a \) and \( \Sigma_s' = \Sigma_s(1 - g) \), where \( \Sigma_a \) and \( \Sigma_s \) are the absorption and scattering cross sections per unit volume. However, the validity of the diffusion approximation of the transport equation is restricted to cases in which the reduced albedo \( c' = \Sigma_s'/\Sigma_a \) is close to 1.

Previous studies, using rigorous solutions of the transport equation for Henyey-Greenstein scattering as well as for Rayleigh-Gans scattering, have confirmed the validity of the diffusion approximation for values of \( c' \) close to 1.0. A more important conclusion from these studies was that light propagation in the whole range of \( 0 < c' < 1 \) proved to be almost insensitive to changes in \( g \) when the values of \( \sigma_a \) and \( \sigma_s' \) are unchanged and \( g \) remains in the range between 0.9 and 1. It has been concluded therefore that \( \sigma_a \) and \( \sigma_s' \) are the properties that mainly influence light propagation within scattering media where \( g \geq 0.9 \). Scatterers within that range of \( g \) include red blood cells and the main part of scatterers within the human skin.

The aim of this study is to investigate the Mie results for the property \( \sigma_s' \) for single spherical particles without absorption as a function of size and relative refractive index. Results are compared with data reported by others. The applicability of the results to spheres with small values of the absorption cross section and to mixtures of spheres of different sizes is discussed. It is assumed throughout that the
scattering process at a single particle is not influenced by other particles. Possible application of the present results to the scattering properties of blood and tissues (e.g., the skin) is also discussed.

Mie Scattering for a Single Sphere

Methods

Values of \( \sigma_s' \) can be derived for single spherical particles that are illuminated by a plane wave by using Mie theory.\(^{10,12} \) The scattered field is determined by the size parameter \( x = 2\pi a/\lambda \), where \( a \) and \( \lambda \) represent the particle radius and the wavelength in the surrounding medium of the particle, and by \( m \), the ratio of the refractive indices of particle and surrounding medium. Absorption by the spherical particle can be considered as well in Mie theory but is not taken into account in this study.

It is well known that the solution of equations for Mie scattering can lead to serious numerical problems, including decreased accuracy and instability, especially when recurrence relations for the Riccati–Bessel functions are not chosen with care.\(^{13,14} \) Therefore, tabulated results of Mie calculations of different origins are compared in this study.

Our results have been obtained with Mie scattering calculations performed by Zijp, using upward recurrence, and by Sloot, using downward recurrence. Zijp used a program based on the equations given by van de Hulst,\(^{10} \) running on a personal computer with a coprocessor. The program of Sloot et al.\(^{15} \) is a modified version of the homogeneous sphere program reported by Bohren and Huffman,\(^{16} \) running on a Cyber 205 computer.

Values of \( \sigma_s' \equiv \sigma_s(1 - g) \) can be determined from \( Q_{\text{sca}} = \sigma_s/\pi a^2 \), a dimensionless representation of the scattering cross section, and \( g \) (Ref. 13):

\[
Q_{\text{sca}} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n + 1)(|a_n|^2 + |b_n|^2),
\]

\[
g = \frac{4}{x^2 Q_{\text{sca}}^2} \sum_{n=1}^{\infty} \left[ \frac{n(n + 2)}{n + 1} \text{Re}(a_{n+1}^* b_n + a_n b_{n+1}^*) + \frac{2n + 1}{n(n + 1)} \text{Re}(a_n b_n^*) \right],
\]

where \( a_n \) and \( b_n \) are complex Mie coefficients and the * denotes complex conjugation. The programs of Sloot and Zijp both calculate \( Q_{\text{sca}} \) from Eq. (1). Values for \( g \) from Eq. (2) are provided only by the program of Zijp. However, it is also possible to determine \( \sigma_s \) and \( g \) from the sum of the angular intensity functions \( i_1(\theta) \) and \( i_2(\theta) \).\(^{10} \)

\[
Q_{\text{sca}} = \frac{1}{x^2} \int_0^{\pi} [i_1(\theta) + i_2(\theta)] \sin(\theta) d\theta,
\]

\[
g = \frac{\int_0^{\pi} [i_1(\theta) + i_2(\theta)] \cos(\theta) \sin(\theta) d\theta}{\int_0^{\pi} [i_1(\theta) + i_2(\theta)] \sin(\theta) d\theta}.
\]

Equations (3) and (4) were applied to the tabulated values of \( i_1(\theta) + i_2(\theta) \), which were calculated for each degree from the scattering functions \( S_1(\theta) \) and \( S_2(\theta) \) (see Ref. 10, p. 125) by the programs of Sloot and Zijp. In this way possible differences caused by the different recurrence relations could be detected. The Simpson rule was used for the numerical integrations.

Angular Intensity Functions

The angular intensity functions \( i_1(\theta) \) and \( i_2(\theta) \) are functions of \( x \) and \( m \). Distributions for the same values of \( x \) and \( m \) have been compared in order to validate our results. Results for \( x = 35 \) and \( m = 1.2 \) are shown in Table I. The values of Ashley and Cobb,\(^{17} \) who used the data of Gumprecht and Sliepcevich,\(^{18} \) deviate from the other values for many angles, whereas our results with the programs of Zijp and Sloot are in agreement with each other.

Scattering Cross Sections and Asymmetry Factors

Values for \( g \) and \( Q_{\text{sca}} \) have been obtained by using Eqs. (1) and (2) with the program of Zijp for values of \( x \) up to 50 and for \( m \) between 1.025 and 1.20. The results are shown in Figs. 1 and 2. The results show the well-known major oscillations of the values of \( Q_{\text{sca}} \) and \( g \) for increasing values of the size parameter.\(^{10,12,19} \) The major oscillations in \( Q_{\text{sca}} \) can be described for \( m = 1 \) with the anomalous diffraction theory as a function of \( 2\pi(m - 1) \).\(^{10} \) Minor oscillations, or ripples, are found when \( m > 1.1 \) and \( x > 15 \).\(^{10,19} \)

Data in the ranges of \( x \leq 50 \) and 1.025 \( \leq m \leq 1.20 \) are shown in Tables II and III. The values for \( Q_{\text{sca}} \) as calculated from Eq. (1) by both programs deviate by

<table>
<thead>
<tr>
<th>( \theta ) (degrees)</th>
<th>Ashley and Cobb(^{17} )</th>
<th>Sloot et al.(^{15} )</th>
<th>Zijp(^{10} )</th>
</tr>
</thead>
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<tr>
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<td>341035</td>
<td>341036</td>
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<td>4245</td>
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<tr>
<td>30</td>
<td>1201</td>
<td>1189</td>
<td>1189</td>
</tr>
<tr>
<td>40</td>
<td>388.5</td>
<td>393.1</td>
<td>393.0</td>
</tr>
<tr>
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<td>159.8</td>
<td>173.6</td>
<td>173.6</td>
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<td>108.5</td>
<td>108.5</td>
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<tr>
<td>70</td>
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<td>92.76</td>
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<td>86.83</td>
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<td>135.7</td>
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<tr>
<td>170</td>
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<td>278.7</td>
<td>278.7</td>
</tr>
<tr>
<td>180</td>
<td>1478</td>
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</tr>
</tbody>
</table>
Table II. \( Q_{\text{ca}} \) from Mie Calculations with Eq. (1) as a Function of \( x \) and \( m \)

<table>
<thead>
<tr>
<th>( x )</th>
<th>( m = 1.025 )</th>
<th>( m = 1.05 )</th>
<th>( m = 1.10 )</th>
<th>( m = 1.20 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>0.02909</td>
<td>0.11822</td>
<td>0.47705</td>
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<tr>
<td>10</td>
<td>0.12283</td>
<td>0.48353</td>
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<td>3.68513</td>
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<tr>
<td>15</td>
<td>0.27565</td>
<td>1.02865</td>
<td>2.92471</td>
<td>2.48889</td>
</tr>
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<td>0.48094</td>
<td>1.66478</td>
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</tr>
<tr>
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<td>0.73015</td>
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<tr>
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</tr>
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</tr>
<tr>
<td>40</td>
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<td>3.31666</td>
<td>1.67479</td>
<td>2.31762</td>
</tr>
<tr>
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<td>1.94734</td>
<td>3.25098</td>
<td>2.03799</td>
<td>2.31562</td>
</tr>
<tr>
<td>50</td>
<td>2.24667</td>
<td>3.01270</td>
<td>2.44616</td>
<td>1.98649</td>
</tr>
</tbody>
</table>

Larger differences are found between the values for \( Q_{\text{ca}} \) obtained by using Eqs. (1) and (3). However, these differences are smaller than 0.24% for the whole range of \( m \) and \( x \) given in Table II. The differences are introduced by the 1° step size for numerical integration, which may be unacceptable for accurate results, especially for the larger values of \( x \) where fast changes in the angular intensity functions occur. Similar differences are found between values for \( g \) obtained by using Eqs. (2) and (4), which remain smaller than 0.026% in the whole range of \( m \) and \( x \) listed in Table III. The values calculated by using Eqs. (1) and (2) are therefore preferred.

The values of \( Q_{\text{ca}} \) and \( g \) of Chu et al.\(^2\) as given by van de Hulst\(^3\) for \( m = 1.2 \) and \( x = 5-25 \), agree with the current results for both \( Q_{\text{ca}} \) and \( g \) as shown in Table IV. The data for \( Q_{\text{ca}} \) of Wickramasinghe\(^21\) for \( x = 5-15 \) and \( m = 1.1 \) and 1.2 also agree with our results. However, the data for \( g \) as given by Wickramasinghe are probably in error, since they deviate seriously from the data of Chu et al.\(^2\) and the data presented here.

Reduced Scattering Cross Sections of a Sphere

The results for the product \( Q_{\text{ca}}(1 - g) \) are given in Fig. 3 and Table V. It is remarkable that the major oscillations, which are present in \( Q_{\text{ca}} \) as well as in \( g \), do not appear in this product. This might be related to the fact that \( Q_{\text{ca}}(1 - g) \) also equals the change in efficiency for radiation pressure, \( Q_{\text{pr}} \), for nonabsorbing particles after scattering:

\[
Q_{\text{pr}} = Q_{\text{ca}} + Q_{\text{ca}}(1 - g). \tag{5}
\]

Some fluctuations occur for \( x < 5 \), which seem to be independent of \( m \). However, the minor oscillations, or ripples, in \( Q_{\text{ca}} \) and \( g \), which are found when \( m > 1.1 \) and \( x > 15 \) and depend on \( m \), do not disappear in \( Q_{\text{ca}}(1 - g) \).\(^{12,19}\) These minor oscillations in \( Q_{\text{ca}}(1 - g) \) may be related to resonant behavior of surface waves.\(^10\) For larger values of \( x \) or \( m \), the amplitude of the ripple structure becomes larger and more complex.\(^19\)

Ashkin and Dziedzic\(^22\) were able to determine experimentally the ripple structure of the efficiency factor

<table>
<thead>
<tr>
<th>( x )</th>
<th>( m = 1.025 )</th>
<th>( m = 1.05 )</th>
<th>( m = 1.10 )</th>
<th>( m = 1.20 )</th>
</tr>
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<td>15</td>
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<tr>
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<td>0.953063</td>
<td>0.873612</td>
</tr>
</tbody>
</table>
for radiation pressure. They applied optical levitation to single oil drops and showed excellent agreement between the changes of the intensity needed for levitation and the ripple structure in $Q_{pr}$ that is predicted by Mie theory.

### Approximations

**Reduced Scattering Cross Sections**

It is possible to fit a large part of the data for $Q_{ss}(1 - g)$ in the range where the influence of the ripple can be neglected:

$$[Q_{ss}(1 - g)]_{pred} = 3.28x^{0.37}(m - 1)^{2.09},$$  

(6)

as shown in Fig. 3. The given coefficients have been derived by trial and error for values of $x$ and $m$ in the range where $g > 0.90$: $5 < x < 50$ and $1 < m < 1.1$. This is the range for scattering in blood and a large part of scatterers within living tissues. The inaccuracy introduced by Eq. (6) is restricted to a few percent. With the aid of Table V different values for the coefficients may be chosen if only a part of the present range has to be described with higher accuracy. Table VI gives the ratio between the predicted values and the results of Mie calculations. Equation (6) shows that $Q_{ss}(1 - g)$ can be split into a refractive-index-dependent factor and a size-dependent factor, as in the Rayleigh–Debye–Gans equations for intensity at a distance $r$ from a sphere with radius $a$,

$$I(0) = \frac{I_0}{2} \frac{x^2}{r^2} \frac{m^2 - 1}{m^2 + 2} \times \left[ \frac{3}{u^3} \left| \sin(u) - u \cos(u) \right|^2 \right] \left[ 1 + \cos^2(\theta) \right].$$  

(7)

with

$$u = 2x \sin(\theta/2).$$  

(8)

In the Rayleigh–Gans approximation, $g$ is independent of $m$ and $Q_{ss}$ is proportional to $(m^2 - 1)/(m^2 + 2)$ or $(m - 1)^3$ if $m \approx 1$. As a consequence, $Q_{ss}(1 - g)$ will be proportional to $(m - 1)^3$. However, in our range of application, $(m - 1)^2$ best describes the refractive-index-dependent term of the Mie results.

The results for Mie calculations have been compared with Rayleigh–Gans calculations, as shown in Fig. 4. Therefore, Eq. (7) was rewritten in the following form:

$$i_i(\theta) + i_o(\theta) = 2 \left( \frac{2\pi r^2 I(0)}{I_0} \right) = x^4 \frac{m^2 - 1}{m^2 + 2} \times \left[ \frac{3}{u^3} \left| \sin(u) - u \cos(u) \right|^2 \right] \left[ 1 + \cos^2(\theta) \right].$$  

(9)

$Q_{ss}$ and $g$ are found by numerical integration with application of Eqs. (3) and (4) and Eq. (9) for 900...
angles. It appears that the Rayleigh–Gans theory predicts \( Q_{\text{Rayleigh–Gans}}(1 - g) \) much better than \( Q_{\text{Mie}} \) or \( g \), since \( Q_{\text{Rayleigh–Gans}} \) is a factor of 4 too large for \( x = 50 \) and \( m = 1.05 \) with Rayleigh–Gans theory, whereas \( Q_{\text{Rayleigh–Gans}}(1 - g) \) is only 20% too large. The behavior for \( x < 5 \) is depicted to advantage by the Rayleigh–Gans theory, as expected.

Another remarkable conclusion can be drawn from Eq. (6): the ratio of the reduced scattering cross sections at two different wavelengths is a function only of the wavelength ratio and the values of \( m \). It can be further reduced to

\[
\frac{\sigma_r'(\lambda_1)}{\sigma_r'(\lambda_2)} = \frac{\lambda_2^{2} (m_1 - 1)^{2} \sin^2 \theta}{\lambda_1^{2} (m_2 - 1)}. \quad (10)
\]

The ratio of the reduced cross sections proves to be independent of the particle size in this range and can thus be predicted with an accuracy of a few percent without knowledge of the particle size. It is clear from Fig. 3 that such a relation cannot be derived in the range where rippling occurs.

Mixtures of Spheres

It is possible to calculate an average value of the reduced scattering cross section for a mixture of spheres, \( \sigma_r'(\text{mixture}) \), from individual values of \( \sigma_r \) and \( g \), if it is assumed that scattering at a single particle is not influenced by other particles and that angular intensities can be added without considering coherence. In this case we may write

\[
I(\theta) = \Sigma \frac{\text{frac}(i) I(\theta, i)}{\Sigma \text{frac}(i) I(\theta, i)}. \quad (11)
\]

where the fractions of the different scatterers should be weighted with their concentrations in the medium. For a mixture of scatterers, the following relation can then be derived:

\[
\sigma_r'(\text{mixture}) = (\sigma_r(1 - g)) = (\sigma_r - (\sigma_r g)) = (\sigma_r(1 - g)). \quad (12)
\]

The last term equals \( (\sigma_r') \). This means that the reduced scattering cross section of a mixture \( \sigma_r' \) (mixture) can be derived from the individual values of \( \sigma_r'(i) \) and their concentrations, without knowledge of the individual values of \( \sigma_r'(i) \) and \( g(i) \).

Substitution of Eq. (6) shows that it is even possible to apply Eq. (10) to mixtures of spheres when the size distribution, concentration, and refractive indices are unknown. However, \( (m_1 - 1)/(m_2 - 1) \) must be available in that case and must have the same value for all spheres. And, of course, \( m(i) \) and \( x(i) \) must be within the range of validity of Eq. (10).

Influence of Absorption when \( \sigma_a \ll \sigma_r \)

When absorption is introduced with \( \sigma_a \ll \sigma_r \), as in the case of red blood cells for 630 nm < \( \lambda \) < 1000 nm, it has been shown by Mie calculations considering absorption that \( \sigma_a + \sigma_r \) is approximately independent of absorption for red blood cells.\(^7\) In that case \( \sigma_r \) can be found by subtraction of \( \sigma_a \) from the value found in Table II. Since it has also been recognized that values of \( g \) are virtually independent of small amounts of absorption,\(^7\) it is possible to predict \( \sigma_r' \). The validity of this correction method is supported by the figures of Irvine\(^9\) for \( m = 1.2 \) and \( x = 0-30 \): it is possible to predict \( \sigma_a + \sigma_r' \) to within 6% as long as tan(\( \beta \)) < 0.10. Herein, \( n' = \gamma \lambda/(4\pi) \), where \( \gamma \) is the absorption coefficient of the material of the sphere, and values for \( \sigma_r \) are calculated with the relations given by Bohren and Nevitt.\(^2\) However, this correction method cannot be used for mixtures where \( x \) and \( m \) are unknown.

When \( x \) and \( m \) are not known and Eq. (10) is to be used, another approach must be found. If \( \sigma_a \ll \sigma_r \), we may neglect the influence of absorption on \( g \) and \( \sigma_r' \) and thus on \( \sigma_r'(\text{mixture}) \). From the figures of Irvine for \( m = 1.2 \) and \( x = 0-30 \), it is found that the deviation of \( \sigma_r + \sigma_r' \) from the Mie results without absorption is < 10% as long as \( n'/m - 1 \leq 0.10 \). \( Q_{\text{abs}} \) is predicted with the anomalous diffraction approximation.\(^10\) For many biomedical applications in which size distributions of particles are not known, this approximation is sufficient.

Biomedical Applications

In this section we discuss whether Eqs. (6) and (10) can be used to describe the scattering properties of red blood cells and several tissues. Relations such as Eqs. (6) and (10) might prove to be important for biomedical applications, especially in noninvasive applications in which light propagation is compared at several wavelengths, as in near-infrared spectroscopy and pulse oximetry. With these methods exact knowledge of phase functions and scattering coefficients is
not available because of the variation of these scattering properties between subjects and even with time, but with Eq. (10) it becomes possible to predict a ratio of reduced scattering coefficients.

Red blood cells are within the range of validity of Eq. (6) for size (5 < x < 50) and refractive index (1 < m < 1.1) for visible and infrared light. However, red blood cells are not spherically shaped, and the distance between adjacent red blood cells in blood vessels is short. Although Mie calculations are successful in predicting experimentally found values for σ'_r of red blood cells, they overpredict experimentally found values of σ'_r by a factor of up to 3 or more. More work is therefore needed to overcome these discrepancies between theory and experiment in order to validate the use of Eq. (6) for nonspherical scatterers such as red blood cells. However, the influence of the nonspherical shape of scatterers and distance to adjacent scatterers on the ratio of reduced scattering cross sections is probably smaller than the influence of shape and distance on the scattering cross sections themselves. This may justify the use of Eq. (10) for red blood cells.

Differences between refractive indices within human tissues are probably small enough to obey 1.0 < m < 1.1. Measurements of the asymmetry factor with a goniometer by Flock et al. at 633 nm for chicken and bovine muscle as well as pig brain were all above 0.94, indicating that the size of most scatterers within these tissues is large enough to fit in the 5 < x < 50 range. Only a small fraction of the scatterers in these tissues might have a smaller size. However, much lower values (0.6 < g < 0.8) were found in tumor and adipose tissue. For human dermis at 633 nm, the intensity distribution after single scattering, as measured by Jacques et al., indicates that 90% of the scattering events can be described with Henyey-Greenstein scattering with an asymmetry factor of 0.92. The remaining scattering events within the dermis samples have been ascribed to scattering by small scatterers.

For tissues with a relatively large amount of small scatterers, the following approach may be chosen. A similar description as with Eq. (10) can be used for the fraction of small scatterers, however, with the wavelength ratio to the power of 4.0 instead of 0.37, according to Rayleigh scattering, and 2.0 instead of 2.09 for the power of the refractive-index-dependent term:

\[
\frac{\sigma'_{s2}(\lambda)}{\sigma'_{s1}(\lambda)} = \frac{|\lambda_2|^{42} m_1 - 1|^{12.6}}{|\lambda_1|^{12.6}}.
\]  

With knowledge of the fraction of σ'_r by small Rayleigh scatterers, f_M, at a given wavelength λ_0 and by applying the mixing rule as given by Eq. (12), we can derive

\[
\frac{\sigma'_r(\lambda)}{\sigma'_r(\lambda_0)} = \frac{\sigma'_{s1}(\lambda)}{\sigma'_{s1}(\lambda_0)} [1 - f_M(\lambda_0)] + \frac{\sigma'_{s2}(\lambda)}{\sigma'_{s2}(\lambda_0)} f_M(\lambda_0),
\]  

where the first term is given by Eq. (10) and the second term by Eq. (13). Figure 5 shows the results for f_M(633) = 0.1, we can expect that Eq. (10) can also be applied with reasonable accuracy to human dermis within the range of red and near-infrared light.

**Conclusions**

The reduced scattering cross section is an important parameter for the description of light propagation in scattering media. This reduced scattering cross section can be predicted to within a few percent for spherical particles in the ranges of 5 < x < 50 and m ≤ 1.10 with the simple equation given [Eq. (6)]. Larger deviations have been observed as soon as the ripple is observed: for m > 1.1 when x ≥ 15. Corrections for small amounts of absorption can be made.

However, it is more important that the ratio of reduced scattering cross sections at different wavelengths can be predicted for a medium with spherical particles without a precisely known size distribution and refractive index as long as x and m are within the range of validity of our equation and scattering at a single particle is not influenced by other particles. The ratio (m_1 - 1)/(m_2 - 1) has to be known. In these conditions, Eq. (10) can be applied to biomedical multiwavelength applications such as whole blood oximetry, pulse oximetry, and tissue spectroscopy.

**References**
