Some analytical approximations to radiative transfer theory and their application for the analysis of reflectance data

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Received 23 November 2007, accepted for publication 17 January 2008
Published 11 February 2008
Online at stacks.iop.org/JOptA/10/035001

Abstract
We derive an analytical approximation in the framework of the radiative transfer theory for use in the analysis of diffuse reflectance measurements. This model uses two parameters to describe a material, the transport free path length, \( l \), and the similarity parameter, \( s \). Using a simple algebraic expression, \( s \) and \( l \) can be applied for the determination of the absorption coefficient \( K_{\text{abs}} \), which can be easily compared to absorption coefficients measured using transmission spectroscopy. \( l \) and \( K_{\text{abs}} \) can be seen as equivalent to the \( S \) and \( K \) parameters, respectively, in the Kubelka–Munk formulation. The advantage of our approximation is a clear basis in the complete radiative transfer theory. We demonstrate the application of our model to a range of different paper types and to fabrics treated with known levels of a dye.

Keywords: light scattering, radiative transfer, spectral reflectance measurements

1. Introduction

The analysis and optimization of the reflective characteristics of a range of materials which exhibit strong multiple scattering is of great importance for a number of technological applications, such as the manufacture of paper and the application of paints and other coatings. This is also the case for spectral transmission in the development of UV sunblocks, for example. Such an analysis is usually performed on the basis of Kubelka–Munk theory (KMT) (Kubelka and Munk 1931). However, there are several shortcomings of KMT. In particular, this theory does not operate directly with standard notions of the radiative transfer theory like the phase function, the single scattering albedo, and the extinction coefficient (Chandrasekhar 1960). Also, KMT is not able to describe the angular dependence of the reflectance. It is a two-flux model, which can be obtained from the radiative transfer equation by assuming an isotropic distribution for the intensity of the multiply scattered diffuse radiation. With this in mind, we design here a simple optical model for multiple scattering materials, which is more accurate than that given by KMT for the small absorption case and has a solid theoretical basis.

The geometrical thickness of most the materials we choose to study exceeds the scattering mean free path \( l_s \) defined as the inverse value of the scattering coefficient \( K_{\text{sca}} \). Due to this fact bright materials are opaque and multiple light scattering dominates. Therefore, light waves perform a random walk with an average step length and velocity. The average step length is given by \( l \). The transport mean free path is defined as the average distance over which a light wave completely loses memory of its original propagation direction. Therefore, \( l \) is the single most important characteristic of any multiply light scattering material.
The similarity parameter, \( s \), is defined as:

\[
s = \sqrt{\frac{1 - s}{1 - s_0}},
\]

where \( s_0 = 1 - \frac{K_{\text{abs}}}{K_{\text{abs}} + K_{\text{sca}}} \) is the single scattering albedo, which equals one for non-absorbing media, and \( g \) is the average cosine of scattering angle. All materials absorb light in various degrees depending on the structure of a given material. Therefore, \( s \) generally differs from zero. \( s \) is a function of wavelength; it determines the strength of light reflection and absorption for a semi-infinite layer. A semi-infinite layer is defined as a layer whose thickness can be increased without changes in its reflective properties. van de Hulst (1974) has shown that the diffuse reflectance of light from a semi-infinite layer under diffuse light illumination conditions is determined almost exclusively by the parameter \( s \). Therefore, only this parameter can be derived from the corresponding measurements.

The main task of this paper is to derive the local optical characteristics of a material such as \( l \), \( s \) and \( K_{\text{abs}} \) from experimental measurements of light reflection or transmission.

First of all we find simple analytical equations for diffuse reflection and transmission coefficients of optically thick layers in the weakly absorbing limit. This can be done as follows. To start with, we take a finite but optically thick layer illuminated by diffuse light and relate its diffuse transmission \( t \) and reflection \( r \) coefficients to the coefficient of diffuse reflection \( r_\infty \) for the case of a semi-infinite layer having the same optical characteristics.

For this, we truncate the semi-infinite layer at an optical thickness, \( r = K_{\text{ext}} z_0 \Rightarrow 1 (K_{\text{ext}} = K_{\text{abs}} + K_{\text{sca}} \) is the extinction coefficient of a layer and \( z_0 \) is the depth; see figure 1). Because the medium as considered in figure 1 is semi-infinite, the values of reflection coefficients for the whole layer starting at \( z = 0 \) and also a sub-layer starting at \( z = z_0 \) coincide. Due to highly developed multiple light scattering in the system under consideration, the two-layer system as shown in figure 1 can be considered as a single optically thick layer of the geometrical thickness \( z_0 \) over an underlying Lambertian surface with albedo equal to \( r_\infty \).

It is easy to show that the diffuse reflection of such a system (equal to \( r_\infty \) by definition) under diffuse light illumination conditions is related to the diffuse reflection of \( r \) of the upper layer by the following equation:

\[
r_\infty = r + \frac{tr_\infty l}{1 - tr_\infty}, \tag{1}
\]

where \( t \) is the diffuse transmittance of the upper layer (see figure 1). The dominator accounts for multiple reflections between the layer and the underlying surface with the Lambertian albedo \( r_\infty \). On the other hand, the value of \( r_\infty \) can be presented as a sum of the reflection \( r \) from the upper layer shown in figure 1 and the contribution \( \Delta r \) from the bottom layer. So we can write

\[
r_\infty = r + \Delta r, \tag{2}
\]

where a simple physical consideration allows us to present \( \Delta r \) as the product of following three processes:

- the transmittance \( t \) by the upper layer;
- the reflectance \( r_\infty \) from the bottom layer;
- the attenuation \( \kappa \) of light reflected from the lower layer during the propagation process in the upper layer.

It is known from the general radiative transfer theory (van de Hulst 1980) that the attenuation \( \kappa \) can be described by the exponent \( \kappa = \exp(-\gamma \tau) \) for the case of optically thick layers as discussed here. So, finally, we have

\[
\Delta r = tr_\infty e^{-\gamma \tau}, \tag{3}
\]

where \( \gamma \) is called the diffusion exponent. The value of \( \gamma \) can be calculated from the corresponding characteristic integral equation as described by van de Hulst (1980); \( \tau \) is the optical thickness, defined above. For the case of weakly absorbing media, \( \gamma \) is given simply by the following equation (van de Hulst 1980):

\[
\gamma = \sqrt{3(1 - s_0)} q. \tag{4}
\]

where \( s_0 = 1 - K_{\text{abs}}/K_{\text{ext}} \) is the probability of photon survival in a single scattering process (close to one for an optically bright material) and \( q = 1 - g \) is the symmetry parameter, which is equal to the ratio \( l_e/l \), where \( l_e = K_{\text{ext}}^{-1} \). It is known that for isotropic scattering (scattering in any direction occurs with the same probability) \( l = l_e \) and, therefore, \( q = 1 \). Materials such as textiles and paper are composed of scatterers which are large compared to the wavelength of incident light. Therefore, the value of \( q \) must be a small number \( (q \ll 1) \) due to the predominant light scattering in the forward direction by particles with sizes much larger than the wavelength of incident radiation.

The comparison of equations (1) and (2) gives

\[
\Delta r = \frac{tr_\infty l}{1 - tr_\infty}. \tag{5}
\]
Comparing this with equation (3), we arrive at the following relationship:
\[ e^{-\gamma \tau} = \frac{t}{1 - r r_{\infty}}, \tag{6} \]
which shows that the transmission \( t \) can be related to the reflection \( r \) using the following equation:
\[ t = (1 - r r_{\infty}) e^{-\gamma \tau}. \tag{7} \]

It follows for non-absorbing media by definition that \( r_{\infty} = 1, \omega_0 = 1, \gamma = 0 \) and, therefore, equation (7) gives \( t = 1 - r \), which is just the energy conservation law for the problem at hand. Also we have, as \( \tau \to \infty, t \to 0 \) as it should be; therefore, equation (7) has correct asymptotic limits.

The substitution of equation (7) into equation (3) gives
\[ \Delta r = (1 - r r_{\infty}) r_{\infty} e^{-2x}, \tag{8} \]
where \( x = \gamma \tau \). We obtain from equations (2) and (8) \( r_{\infty} = r + (1 - r r_{\infty}) r_{\infty} e^{-2x} \) and, therefore,
\[ r = \frac{r_{\infty} (1 - e^{-2x})}{1 - r_{\infty}^{-2} e^{-2x}}. \tag{9} \]

This equation can be used for the determination of \( x \) from measurements of the pair \((r, r_{\infty})\). This also offers a possibility to determine the diffusion exponent \( \gamma \), if \( \tau \) is measured independently (\( \gamma = x/\tau \)).

It follows from equations (7) and (9) that
\[ t = \frac{(1 - r_{\infty}^{-2}) e^{-x}}{1 - r_{\infty}^{-2} e^{-2x}}. \tag{10} \]

Therefore, we have reached our goal and related the pair \((r, t)\) to the value of \( r_{\infty} \) (see equations (9) and (10)). Interestingly, the pair \((r, t)\) is completely defined, if values \((x, r_{\infty})\) are known.

Now we can use the well-known result of exact radiative transfer theory valid as \( \omega_0 \to 1 \) (van de Hulst 1980):
\[ r_{\infty} = 1 - a s, \tag{11} \]
where \( a = 4/\sqrt{3} \). This allows us to relate pairs \((r, t)\) and \((x, s)\) analytically.

It appears that equation (11) is only valid for values of \( \omega_0 \geq 0.999 \), which is too restrictive for our applications. So we need to extend the range of validity of this equation. This can be done as follows. We represent \( r_{\infty} \) as series with respect to \( \omega_0 \). Then it follows that
\[ r_{\infty} = \sum_{j=0}^{\infty} \sigma_j \omega_0^j, \tag{12} \]
where \( \sigma_j \) are unknown coefficients. This series is poorly convergent as \( \omega_0 \to 1 \). Therefore, we need to introduce the probability of photon absorption \( \beta = 1 - \omega_0 \) in equation (12). Then it follows that
\[ r_{\infty} = \sum_{j=0}^{\infty} \sigma_j (1 - \beta)^j \tag{13} \]
or
\[ r_{\infty} = 1 - (j) \beta + \frac{(j(j - 1)) \beta^2}{2} - \ldots, \tag{14} \]
where we introduced the following notation: \( (j) \equiv \sum_{j=0}^{\infty} j \sigma_j \), \( (j(j - 1)) \equiv \sum_{j=0}^{\infty} j(j - 1) \sigma_j \), etc. Clearly, the largest contribution to \( r_{\infty} \) comes from large values of \( j \). Therefore, we may assume that \( (j(j - 1)) \approx (j^2) \) and similar for higher-order terms. This allows us to derive
\[ r_{\infty} = 1 - (j) \beta + \frac{(j^2) \beta^2}{2} - \ldots = e^{-N \beta}. \tag{15} \]

The constant \( N \) can be found from the comparison of equation (15) as \( \beta \to 0 (r_{\infty} = 1 - N \beta) \) with equation (11). In particular, it follows that
\[ N = \frac{as}{\beta}, \tag{16} \]
and, therefore,
\[ r_{\infty} = e^{-y}, \tag{17} \]
where \( y = as; s \) is the similarity parameter. This means that the similarity parameter \( s \) can be determined from measurements of \( r_{\infty} \).

Finally, we can write for the pair \((t, r)\) using equations (9), (10), (17):
\[ t = \frac{e^{-y} - e^{-2x-y}}{1 - e^{-2x-2y}}, \tag{18} \]
\[ l = \frac{e^{-x} - e^{-2x-y}}{1 - e^{-2x-2y}}. \tag{19} \]

So we have obtained the relationships of diffuse reflectance and transmittance with local optical characteristics of the medium such as the pair \((x, y)\) or \((l, s)\). It follows for weakly absorbing media that \( x/y = 0.75L/l \), where \( L \) is the thickness of the scattering layer. Therefore, values of \( l \) and \( s \) can be determined from reflectance and transmittance measurements. Also, one can easily determine the absorption coefficient to be \( K_{abs} = xy/4L = s^2/l \).

It follows from equations (2), (3), (17) that \( r \) can be represented by
\[ r = r_{\infty} - te^{-x-y}. \tag{20} \]

This equation holds for diffuse light illumination and observation conditions. Let us imagine that a layer is illuminated by a monodirectional beam along the direction \( \theta_0 \) from the normal to the layer. Then one obtains instead of equation (20)
\[ r_d (\xi) = r_{d\infty} (\xi) - t_d (\xi) e^{-x-y}, \tag{21} \]
where \( \xi = \cos \theta_0 \), \( r_d (\xi) \) is the diffuse reflectance under monodirectional illumination by a wide light beam, and \( t_d (\xi) \) is the diffuse transmittance under the same illumination conditions. It is known (Zege et al 1991, Kokhanovsky et al 2004) that \( r_{d\infty} (\xi) = r_{\infty}^d (\xi) \) and \( t_d (\xi) = tu (\xi) \), where \( u (\xi) \) is the angular distribution of light in the Milne problem, i.e., for light
escaping a semi-infinite non-absorbing layer with sources of radiation placed at infinity inside the medium. This function can be approximated as (Kokhanovsky 2004)

\[ u(\xi) = \frac{\lambda}{\xi}(1 + 2\xi), \]

independent of the specific scattering law. Due to the reciprocity principle equation, (21) also holds for diffuse light illumination conditions and observation along the direction specified by the angle \( \theta_0 \).

Clearly, we have for the reflection function instead of equation (21):

\[ R(\xi, \eta, \varphi) = R_\infty(\xi, \eta, \varphi) - T(\xi, \eta)e^{-\xi-\varphi}, \]

where the reflection function \( T(\xi, \eta) \) is given by the following formula: \( T(\xi, \eta) = tu(\xi)u(\eta) \) (due to the reciprocity principle) and (Zege et al. 1991, Kokhanovsky 2004) \( R_\infty(\xi, \eta, \varphi) = R_\infty(\xi, \eta, \varphi) \).

In the next section we will apply these formulae to derive some important local optical characteristics of materials from routine reflection or transmission measurements for different observation and illumination conditions. These parameters can be used to predict the reflective and transmittive properties of a given material for arbitrary values of \( L \). The equations given here are well known in the remote sensing literature (Zege et al. 1991, Kokhanovsky 2004). In the next section we will apply these formulae to derive some important local optical characteristics of paper and fabric using diffuse reflectance spectroscopy; the same analysis could be applied to any other multiple scattering substrate. In particular, the case of measurements at a fixed observation angle for diffuse illumination conditions will be studied. Then we need to use equation (21), substituting \( \xi \) by \( \eta \).

The accuracy of equation (21) compared to the solution of the radiative transfer equation (Chandrasekhar 1960) is given in figure 2 for the exactly solvable case of a plane-parallel layer with polydispersed spherical water droplets. We have assumed that the wavelength \( \lambda = 550 \text{ nm} \) and the observation angle \( \theta = 8^\circ \). It was assumed that droplets are characterized by the gamma particle size distribution \( f(a) = \frac{Aa^k}{\Gamma(k+1)}\exp(-1.5a) \), where \( A \) is the normalization constant. In this case, \( g = 0.85 \). We see that the error is smaller than 2% at \( r_\delta > 0.5 \), which is quite suitable for retrievals performed in the next section (for bright textiles and papers), especially taking into account various uncertainties inherent to optics of textiles and papers (e.g., possible close-packed media effects). Calculations were carried out for different values of the single scattering albedo \( \omega_0 \) (although in reality water is non-absorbing in the visible so \( \omega_0 = 1 \)). In calculations for the case of non-absorbing media, the limit of equation (21) as \( \omega_0 \to 1 \) was used:

\[ r_\delta(\eta) = 1 - tu(\eta), \]

where

\[ t = \frac{1}{1 + 0.75\tau(1-g)}. \]

The accuracy at small values of \( \tau \) can be increased using the constant 1.072 instead of 1.0 in the first term of the denominator of equation (25) (Kokhanovsky 2004).

3. Experiment

Measurements of the diffuse reflectance, \( r_\delta(\theta) \), for a variety of different types of paper (lens tissue, manila, white tissue and blue tissue) and fabrics dyed with different levels of a dye were performed using a Hunterlab Ultrascan Pro diffuse reflectance spectrometer. A diffuse illumination–monodirectional observation scheme was used. Samples of interest are presented to a 25.4 mm port in the side of an ‘integrating sphere’, and the sample is folded so as to obtain a series of different thicknesses corresponding to 1, 2, 4, 8, 16 and 32 times the thickness of a single sheet of the sample. The sample is backed with a light trap, i.e. with zero reflectance. The illumination simulates the International Commission on Illumination defined D65 illuminant. Data are collected over the wavelength range 400–1000 nm. The collection angle, \( \theta = 8^\circ \), and in this instance the specular reflection is included. Since, in this instance, cos \( \theta \) is close to 1 we conclude that the factor \( u \) defined above (see equation (22)) is close to 9/7.

The fabric samples were a set of woven cotton samples treated with the dye Direct Violet 51. Six samples were measured in this sequence—the new fabric, fabrics with dye at 2, 4, 8 and 16 ppm and a new fabric treated in the same solution as that used to carry the dye, but without the dye (control).

For the fabric samples the reflectances lie in the range 0.7–0.95 in the visible region of the spectrum, whilst for the paper samples the reflectance varies significantly across the spectrum. Examples of the measured ‘semi-infinite’
reflectance for the set of papers as a function of the wavelength are shown in figure 3(a); by semi-infinite we mean the reflectance for 32 layers of the paper. These samples demonstrate a range of different behaviors: manila shows a high reflectance at long wavelengths with steadily decreasing reflectance at shorter wavelengths; lens tissue exhibits large reflectance across most of the spectrum with some reduction towards shorter wavelengths; white tissue exhibits high reflectance across most of the spectrum with a peak around 450 nm and a trough at the shortest wavelengths measured—this is characteristic of the presence of a fluorescent whitening agent (FWA). Finally, the blue tissue exhibits reflectance properties broadly similar to that of lens tissue, but with the addition of a broad trough corresponding to absorption by the dye which makes it blue.

The change in diffuse reflectance $r_d$ as a function of thickness is shown in figure 3(b) for this set of different papers. The single layer thicknesses for the samples are as follows: blue tissue, 0.18 mm; manila, 0.088 mm; white tissue, 0.059 mm; and lens tissue, 0.033 mm. The manila and white tissue exhibit clear plateaus in the diffuse reflectance, whilst the other paper types exhibit some flattening as a function of increased thickness.

We have fitted equation (21) to diffuse reflectance data measured as a function of thickness to obtain the transport free path length, $l$, and the similarity parameter, $s$, from which can be derived the absorption coefficient, $K_{abs} = xy/4L = s^2/l$. Fitting was limited to sample measurements where $r_d > 0.5$; this limit is chosen a little arbitrarily as the point at which the approximation deviates from the full theory by 2% (see...
Figure 5. (a) Dependence of the ‘semi-infinite’ diffuse reflectance on wavelength for fabrics dyed with Direct Violet 51 at different levels. (b) Dependence of absorption coefficient on wavelength for fabrics dyed at different levels with Direct Violet 51. (c) Dependence of transport free path length on wavelength for fabrics dyed at different levels with Direct Violet 51.

Fitting was done using the Nelder–Mead simplex method, implemented in Matlab. Applying this analysis to five repeated measurements of a single sample we find a standard deviation of 0.3% in the diffuse reflectance, 3.5% standard deviation in the similarity parameter, and 2% standard deviation in the transport free path length.

Rather than using the similarity parameter directly, we will present the absorption coefficient, \( K_{\text{abs}} = s^2/l \). The \( K_{\text{abs}} \) spectrum and the \( l \) spectrum are shown in figure 4. The manila exhibits relatively large absorption across the visible range, increasing for shorter wavelengths. Blue tissue exhibits a clear absorption peak at 600 nm (i.e. in the red region of the spectrum), just as we would expect for a blue object. The three tissue samples all exhibit an increase in absorption at the shortest wavelengths. Since the white tissue clearly contains a fluorescent agent, the results of the fit over the region of the fluorescent emission band, typically \( \sim 400-500 \) nm, are invalid since the model does not take account of fluorescent emission.

The value of transport mean free path, \( l \), is in the range 0.03–0.07 mm; paper samples with a higher mass density have a smaller transport mean free path, as we would expect since decreasing \( l \) corresponds to an increase in scattering. \( l \) depends only weakly on the wavelength; this is due to the fact that scatterers in paper are much larger than the wavelength of the incident light.

The diffuse reflectance for the dyed fabrics is shown in figure 5(a); a similar analysis to obtain \( l \) and \( K_{\text{abs}} \) is applied, the results of which are shown in figures 5(b) and (c). The transport free path length, \( l \), is close to constant across the spectral range with small deviations from the trend value at the location of the peak in the absorption coefficient \( K_{\text{abs}} \); for the highest concentration of dye this amounts to \( \sim 5\% \) deviation from the
trend. There are two possible causes for this deviation: it may be purely numerical, i.e. a result of the fitting process, or there may be a physical mechanism, which means that the transport free path length does change on the addition of dye.

Clearly, as we would expect for a visible dye, there is a significant dependence of the absorption coefficient on the wavelength. \( K_{abs} \) exhibits a strong peak with a maximum at 575 nm.

We can convert \( K_{abs} \), measured from the diffuse reflectance to a specific absorption coefficient, \( \varepsilon_R \), as follows:

\[
\varepsilon_R = \nu K_{abs}/c, \tag{26}
\]

where \( c \) is the dye concentration and \( \nu = \log_{10}(e) \). The multiplication by \( \nu \) is required to account for slightly differing unit definitions in the reflectance and transmission measurements. This specific absorption coefficient, \( \varepsilon_R \), is directly comparable to the specific absorption coefficient, \( \varepsilon_s \), of the dye measured in solution using a UV–vis spectrophotometer. Figure 6 shows \( \varepsilon_R \) for the fabric loaded with 16 ppm Direct Violet 51 along with \( \varepsilon_s \), measured in solution. The spectral shapes for these spectra are very similar in shape and intensity. However, in solution the absorption maximum occurs at 550 nm. This type of shift has been observed previously and can be attributed to changes in the chemical environment of the dye which change its absorption properties (Abbott et al 2004).

Figure 7 shows that the value of \( K_{abs} \) is proportional to the dye concentration. This confirms the theoretical dependence given above. Indeed, it was stressed above that \( s \propto \sqrt{1 - \alpha} \), or \( s \propto \sqrt{K_{abs}/K_{ext}} \). We can represent \( K_{abs} \) as:

\[
K_{abs} = K_{ext}^f + K_{abs}^d, \tag{27}
\]

where \( f \) denotes a fabric and \( d \) denotes a dye.

Taking into account that absorption is weak and scattering is strong, we may write \( K_{ext} \approx K_{ext}^f \). This means that

\[
s \propto \sqrt{K_{abs}^f/K_{ext} + K_{abs}^d/K_{ext}}. \tag{28}
\]

It is reasonable to assume that \( K_{abs}^d \sim c \alpha \), where \( \alpha \) is the dye bulk absorption coefficient (Kokhanovsky 2004). So we expect that \( s^2 \) must be proportional to \( c \) because \( K_{abs}^f \ll K_{ext}^f \) in the spectral range studied. This proportionality is confirmed by figure 7. The specific absorption coefficient, \( \varepsilon_T \), measured using UV–vis spectrophotometry at the peak in the absorption spectra is \( 4.69 \times 10^4 \text{ cm}^2 \text{ g}^{-1} \). This compares well with a value of \( \varepsilon_R = 3.93 \times 10^4 \text{ cm}^2 \text{ g}^{-1} \) obtained from the slope of figure 7 multiplied by \( \log_{10}(e) \).

4. Conclusions

We have used simple analytical equations for the analysis of reflection and transmission properties of bright multiply scattering materials. The accuracy of the applied equations for the plane albedo, \( \alpha \), is better than 2% for reflectances above 50%, which is the case for bright fabrics and papers. The transport mean free path, \( l \), was found to be in the range 0.03–0.07 mm at the wavelength 600 nm for the papers studied, with only minor spectral changes in the spectral region 400–1000 nm. The similarity parameter, \( s \), and the transport free path length, \( l \), measured here, can be used to model light propagation in fabrics and papers for a wide range of different illumination and observation conditions.

Acknowledgments

The authors are grateful to E P Zege, I L Katsev, and R Treloar for important discussions on the optics of light scattering media.

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