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Diffusion of light in turbid media with internal reflections

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We relate the the Kubelka-Munk equations for the description of the intensity transfer of light in turbid media to a one-dimensional diffusion equation, which is obtained by averaging the three-dimensional diffusion equation over the lateral directions. This enables us to identify uniquely the Kubelka-Munk parameters and derive expressions for diffuse reflection and transmission coefficients including the effect of internal reflections. Without internal reflections we recover the Kubelka-Munk formulae for these coefficients. We show that the Kubelka-Munk equations are the proper radiative-transfer equations for the one-dimensional diffusion problem.

Key words: *diffusion, turbid media, transfer equations*

1. Introduction

Investigating the reflectance and transmission of turbid media is a widely-used tool for materials characterization with applications ranging from soil science, over medicine, the production of paper and paint, to the design of laser car headlights [1–5]. In the analysis of the observed spectra the theory of diffuse reflectance and transmissance of Kubelka and Munk [6–8], has been widely used. The microscopical significance of the phenomenological parameters S and K appearing in this theory was discussed in many treatments [1, 9–18], but with differing results for these coefficients.

Here we show that for a geometry of rectangular incidence onto a turbid material, in which the scattering is strong enough to lead to diffusive motion of the light intensity, the Kubelka-Munk equations are equivalent to the one-dimensional projection of the 3-dimensional diffusion equation of the light intensity in the medium. This is done in the second section. In the third section we derive expressions for the diffuse reflectance and transmission coefficients, including the effect of internal reflection. The standard Kubelka-Munk results without internal reflection [6, 7] are recovered. In the fourth section we show that the Kubelka-Munk equations are, in fact, the proper radiative-transfer equations for the quasi-onedimensional scattering problem. In the fifth section some conclusions are drawn.

2. Diffusion and Kubelka-Munk equations

In the diffusion approximation [9, 19] the light intensity $U(\mathbf{r})$ and the current density $\mathbf{j}(\mathbf{r})$ obey the steady-state energy-balance and Fick equations

$$\begin{aligned} \nabla \mathbf{j}(\mathbf{r}) &= -\lambda_a U(\mathbf{r}) + \mathcal{J}(\mathbf{r}), \\ \nabla U(\mathbf{r}) &= -\frac{1}{\widetilde{D}} \mathbf{j}(\mathbf{r}), \end{aligned} \tag{1}$$

which are equivalent to the (steady-state) diffusion equation

$$\lambda_a U(\mathbf{r}) = \widetilde{D}\nabla^2 U(\mathbf{r}) + \mathcal{J}(\mathbf{r}). \tag{2}$$

Here, $\mathcal{J}(\mathbf{r})$ is a source term.

The quantity $\widetilde{D}$, which is the diffusivity divided by the light velocity in the material[1] $v = c/n$ is given by [20]

$$\widetilde{D} = D/v = \frac{1}{\lambda_a + 3\lambda_t}, \tag{3}$$

λ_a, λ_s and λ_t are the inverse mean free paths due to absorption, scattering and transport. The latter two are related as

$$\lambda_t = \lambda_s(1 - \langle\cos\gamma\rangle), \tag{4}$$

where γ is the scattering angle and $\langle\cos\gamma\rangle$ is the anisotropy parameter.

The relation of the diffusivity to the absorption parameter λ_a, equation (3) had been subject to a dispute in the literature. It was argued [21–23] that the time-dependent diffusion equation[2]

$$\left(\frac{\partial}{\partial\tau} + \lambda_a\right) U(\mathbf{r},\tau) = \widetilde{D}\nabla^2 U(\mathbf{r},\tau) + \mathcal{J}(\mathbf{r}), \tag{5}$$

with a diffusivity that depends on λ_a, violates the scaling property, obeyed by the radiative transfer equation, that the solution of the equation in the presence of absorption should be of the form

$$U(\mathbf{r},\tau) = \mathrm{e}^{-\lambda_a\tau} U_0(\mathbf{r},\tau), \tag{6}$$

where $U_0(\mathbf{r},\tau)$ is the solution of the equation with $\lambda_a = 0$. Therefore, it was argued in [21–23] that the diffusivity should not depend on the absorptivity λ_a. The counter argument is that the proper generalization of the steady-state diffusion equation (2) is *not* equation (5), but a damped telegrapher's equation [19, 20], which obeys the proper scaling. However, this equation should reduce to the wave equation of light for short times [20]. This condition enforces the form (3) of the diffusivity, rather than the form $\widetilde{D} = [3(\lambda_a + \lambda_t)]^{-1}$ according to the conventional literature (e.g. [9]). We repeat Durian's [20] argument in the Appendix.

Let us now consider the geometry of a diffusive-reflection (or -transmission) setup with uniform illumination, i.e., an incoming plane wave in the z direction onto a sample with surface at the $z = 0$ plane, thickness t in z direction and a large incidence area $A \to \infty$ in (x, y) direction (see figure 1).

Instead of considering a three-dimensional diffusion problem, in which the the material parameters are assumed to depend only on the z direction, as usually done [9, 24], we consider the photon density $\bar{U}(z)$, photon current $\bar{j}(z)$, and source function $\bar{\mathcal{J}}(z)$, averaged over the lateral (x, y) directions:

$$\begin{aligned} \bar{U}(z) &= \frac{1}{A}\int_A \mathrm{d}x\,\mathrm{d}y\, U(\mathbf{r}), \qquad \bar{j}(z) = \frac{1}{A}\int_A \mathrm{d}x\,\mathrm{d}y\, j_z(\mathbf{r}), \\ \bar{\mathcal{J}}(z) &= \frac{1}{A}\int_A \mathrm{d}x\,\mathrm{d}y \mathcal{J}(\mathbf{r}). \end{aligned} \tag{7}$$

It is evident that these quantities obey the following (quasi-) one-dimensional equations

$$\begin{aligned} \frac{\partial}{\partial z}\bar{j}(z) &= -\lambda_a \bar{U}(z) + \bar{\mathcal{J}}(z), \\ \frac{\partial}{\partial z}\bar{U}(z) &= -\frac{1}{\widetilde{D}}\bar{j}(z), \end{aligned} \tag{8}$$

which lead to the one-dimensional diffusion equation

$$\lambda_a \bar{U}(z) = \widetilde{D}\frac{\partial^2}{\partial z^2}\bar{U}(z) + \bar{\mathcal{J}}(z). \tag{9}$$

[1] c is the light velocity and n is the index of refraction.
[2] $\tau = vt$ is the velocity-scaled time.

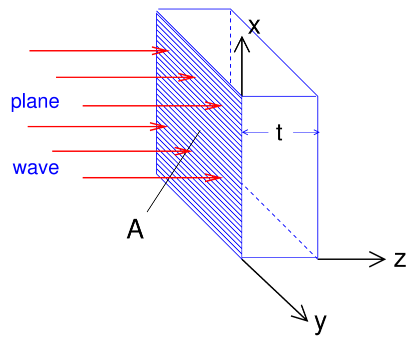

Figure 1. (Colour online) Geometry for the discussion of diffuse reflectance and transmission with uniform illumination (plane-wave incidence). We consider a slab of thickness t, which is infinitely extended in x and y direction.

Defining now the incoming and outgoing currents as

$$I_{\pm}(z) = \frac{1}{2}\left[\bar{U}(z) \pm \bar{j}(z)\right], \tag{10}$$

we obtain from the diffusion equations (8) the Kubelka-Munk equations

$$\begin{aligned} \left(\frac{\partial}{\partial z} + K\right) I_+(z) &= -S\big(I_+(z) - I_-(z)\big) + \bar{\mathcal{J}}(z), \\ \left(-\frac{\partial}{\partial z} + K\right) I_-(z) &= -S\big(I_-(z) - I_+(z)\big) + \bar{\mathcal{J}}(z), \end{aligned} \tag{11}$$

with

$$\begin{aligned} K &= \lambda_a, \\ S &= \frac{1}{2}\left(\frac{1}{\widetilde{D}} - \lambda_a\right) = \frac{3}{2}\lambda_t, \end{aligned} \tag{12}$$

equation (12) can also be written as

$$\frac{1}{\widetilde{D}} = K + 2S. \tag{13}$$

3. Derivation of reflectance and transmission coefficients

Instead of solving equations (11) we solve the diffusion equation (9).

The general solution of the homogeneous diffusion equation [setting $\bar{\mathcal{J}} = 0$ in equation (9)] is

$$\bar{U}(z) = A\mathrm{e}^{\alpha z} + B\mathrm{e}^{-\alpha z}, \tag{14}$$

with the inverse diffusion length

$$\alpha = \sqrt{K/\widetilde{D}} = \sqrt{K(K+2S)}. \tag{15}$$

From the solution (14) we get the incoming and outgoing currents [25, 26]

$$I_{\pm}(z) = \frac{1}{2}\left[A(1 \mp \beta)\mathrm{e}^{\alpha z} + B(1 \pm \beta)\mathrm{e}^{-\alpha z}\right], \tag{16}$$

with

$$\beta = \widetilde{D}\alpha = \sqrt{K\widetilde{D}} = \sqrt{K/(K+2S)}. \tag{17}$$

3.1. Optically thick samples

3.1.1. No reflection at $z = 0$

The appropriate boundary conditions corresponding to optically thick samples without reflection at $z = 0$ are as follows:

$$I_+(0) = \bar{U}_0, \qquad \bar{I}_+(\infty) = 0. \tag{18}$$

The second boundary condition implies $A = 0$. The incoming and outgoing currents are therefore as follows:

$$I_\pm(z) = \frac{1}{2}B(1 \pm \beta)\mathrm{e}^{-\alpha z}. \tag{19}$$

From the first boundary condition we obtain

$$B = \bar{U}_0 \frac{2}{1+\beta}, \tag{20}$$

from which we obtain the incoming current at $z = 0$

$$I_-(0) = \bar{U}_0 \frac{1-\beta}{1+\beta} \tag{21}$$

and hence the reflectivity

$$R_\infty = \frac{I_-(0)}{I_+(0)} = \frac{1-\beta}{1+\beta}. \tag{22}$$

For the Kubelka-Munk function we obtain, using equation (12)

$$\frac{S}{K} = \frac{1}{2}\left[\left(\frac{1+R_\infty}{1-R_\infty}\right)^2 - 1\right] = \frac{2R_\infty}{(1-R_\infty)^2} = \frac{3}{2}\frac{\lambda_t}{\lambda_a}. \tag{23}$$

3.1.2. Reflection at $z = 0$

The first boundary condition is now

$$I_+(0) = U_0 + R_0 I_-(0), \tag{24}$$

where R_0 is the reflectivity at the $z = 0$ boundary. Inserting the expressions (19) for $I_\pm(0)$ we get

$$\frac{2}{1+\beta}I_+(0) = B = R_0\frac{2}{1+\beta}I_+(0) = \frac{2}{1+\beta}U_0 + R_0 R_\infty B \tag{25}$$

from which it follows

$$B = \frac{1+\beta}{2}U_0\frac{1}{1-R_0R_\infty}. \tag{26}$$

The current in reverse direction is given by

$$I_\leftarrow = (1-R_0)I_-(0) \tag{27}$$

and hence

$$R = \frac{1}{U_0}I_\leftarrow = R_\infty\frac{1-R_0}{1-R_0R_\infty}. \tag{28}$$

3.2. Optically thin samples

For optically thin samples with reflectivity R_1 at the back ($z = t$) of the sample and Reflectivity R_0 at the front ($z = 0$) of the sample we have the boundary conditions

$$I_+(0) = \bar{U}_0 + R_0 I_-(0), \qquad I_-(t) = R_1 I_+(t). \tag{29}$$

Using the definition of R_∞, equation (22), we get from the boundary conditions a linear set of equations for the coefficients A and B

$$\frac{2}{1+\beta} I_+(0) = R_\infty A + B = \frac{2}{1+\beta}\left[\bar{U}_0 + R_0 I_-(0)\right] = \frac{2}{1+\beta}\bar{U}_0 + R_0\left[A + BR_\infty\right], \tag{30}$$

which can be put into the form

$$\begin{pmatrix} R_\infty - R_0 & 1 - R_\infty R_0 \\ (1 - R_\infty R_1)\mathrm{e}^{\alpha t} & (R_\infty - R_1)\mathrm{e}^{-\alpha t} \end{pmatrix}\begin{pmatrix} A \\ B \end{pmatrix} = \frac{\mathcal{J}_1}{\beta}\begin{pmatrix} \frac{2}{1+\beta}\bar{U}_0 \\ 0 \end{pmatrix}. \tag{31}$$

The determinant of the coefficient matrix is

$$D = (R_\infty - R_0)(R_\infty - R_1)\mathrm{e}^{-\alpha t} - (1 - R_\infty R_0)(1 - R_\infty R_1)\mathrm{e}^{\alpha t}. \tag{32}$$

Thus, we get from Kramer's rule

$$A = \frac{\bar{U}_0}{D}\frac{2}{1+\beta}\mathrm{e}^{-\alpha t}(R_\infty - R_1), \tag{33}$$

$$B = -\frac{\bar{U}_0}{D}\frac{2}{1+\beta}\mathrm{e}^{\alpha t}(1 - R_1 R_\infty). \tag{34}$$

We obtain for the currents at $z = 0$ and at $z = t$:

$$I_-(0) = \frac{1+\beta}{2}\left[A + R_\infty B\right] = \frac{\bar{U}_0}{D}\left[\mathrm{e}^{-\alpha t}(R_\infty - R_1) - R_\infty \mathrm{e}^{\alpha t}(1 - R_1 R_\infty)\right], \tag{35}$$

$$I_+(t) = \frac{1+\beta}{2}\left[R_\infty A\mathrm{e}^{\alpha t} + B\mathrm{e}^{-\alpha t}\right] = \frac{\bar{U}_0}{D}\left[R_\infty^2 - 1\right], \tag{36}$$

from which we ge the reflectivity R

$$\begin{aligned} R &= \frac{I_\leftarrow}{\bar{U}_0} = (1 - R_0)\frac{I_-(0)}{\bar{U}_0} \\ &= (1 - R_0)R_\infty \frac{\mathrm{e}^{\alpha t}(1 - R_\infty R_1) - \mathrm{e}^{-\alpha t}(1 - R_1/R_\infty)}{(1 - R_\infty R_0)(1 - R_\infty R_1)\mathrm{e}^{\alpha t} - (R_\infty - R_0)(R_\infty - R_1)\mathrm{e}^{-\alpha t}} \end{aligned} \tag{37}$$

and the transmittivity T

$$T = \frac{I_+(t)}{\bar{U}_0} = \frac{1 - R_\infty^2}{(1 - R_\infty R_0)(1 - R_\infty R_1)\mathrm{e}^{\alpha t} - (R_\infty - R_0)(R_\infty - R_1)\mathrm{e}^{-\alpha t}}. \tag{38}$$

Introducing the Kubelka-Munk parameters

$$a = \frac{1}{2}\left(\frac{1}{R_\infty} + R_\infty\right), \quad b = \alpha/S = \frac{1}{2}\left(\frac{1}{R_\infty} - R_\infty\right), \tag{39}$$

we get

$$R = (1 - R_0)\frac{R_1 b\cosh(\alpha t)R_1 b + (1 - R_1 a)\sinh\alpha t}{b(1 - R_0 R_1)\cosh\alpha t + \left[a(1 - R_0 R_1) - R_0 - R_1\right]\sinh\alpha t}, \tag{40}$$

$$T = \frac{b}{b(1 - R_0 R_1)\cosh\alpha t + \left[a(1 - R_0 R_1) - R_0 - R_1\right]\sinh\alpha t}. \tag{41}$$

If we set $R_0 = 0$, we get the formulae of Kubelka [7]

$$R = \frac{1 - R_1 a + R_1 b \coth\alpha t}{a - R_1 + b\coth\alpha t} \tag{42}$$

and

$$T = \frac{b}{b\cosh\alpha t + (a - R_1)\sinh\alpha t}. \tag{43}$$

For $R_0 = R_1 = 0$ we get the standard Kubelka-Munk formulae [7, 25, 26], which do not contain the effect of internal reflections.

$$R = \frac{e^{\alpha t} + e^{-\alpha t}}{e^{\alpha t}\dfrac{1}{R_\infty} - e^{-\alpha t} R_\infty} = \frac{\sinh\alpha t}{a\sinh\alpha t + b\cosh\alpha t}, \tag{44}$$

$$T = \frac{\dfrac{1}{R_\infty} - R_\infty}{e^{\alpha t}\dfrac{1}{R_\infty} - e^{-\alpha t} R_\infty} = \frac{b}{a\sinh\alpha t + b\cosh\alpha t}. \tag{45}$$

Another interesting limit is that of very small R_∞, i.e., $R_\infty \to 0$:

$$R = \frac{R_1 e^{-\alpha t}}{e^{\alpha t} - R_0 R_1 e^{-\alpha t}} = \frac{R_1 e^{-2\alpha t}}{1 - R_0 R_1 e^{-2\alpha t}}, \tag{46}$$

$$T = \frac{1}{e^{\alpha t} - R_0 R_1 e^{-\alpha t}} = \frac{e^{-\alpha t}}{1 - R_0 R_1 e^{-2\alpha t}}. \tag{47}$$

4. Kubelka-Munk equations as one-dimensional radiative-transfer equations

We now want to demonstrate that the Kubelka-Munk equations (11) are the proper radiadive-transfer equations for the diffuse-reflection geometry depicted in figure 1.

We recall the three-dimensional radiative transfer equations of the light intensity in a turbid medium

$$\begin{aligned} \left[\lambda_a + \mathbf{s}\cdot\nabla\right] I(\mathbf{r},\mathbf{s}) &= -\sum_{\mathbf{s}'} q_{\mathbf{s}\mathbf{s}'}\big(I(\mathbf{r},\mathbf{s}) - I(\mathbf{r},\mathbf{s}')\big) \\ &= -\lambda_s I(\mathbf{r},\mathbf{s}) + \sum_{\mathbf{s}'} q_{\mathbf{s}\mathbf{s}'} I(\mathbf{r},\mathbf{s}'), \end{aligned} \tag{48}$$

$I(\mathbf{r},\mathbf{s})$ is the distribution density of light rays passing through $\mathbf{r}$ with the direction $\mathbf{s} = \mathbf{k}/k$, where $\mathbf{k}$ is the wave vector. $q_{\mathbf{s}\mathbf{s}'} = |f(\mathbf{s},\mathbf{s}')|^2$ is the phase function, i.e., the scattering cross-section from $\mathbf{s}$ to $\mathbf{s}'$ with $f(\mathbf{s},\mathbf{s}')$ being the corresponding amplitude. $\sum_{\mathbf{s}'}$ is an integral over the entire solid angle, with the original direction $\mathbf{s}$ being excluded. The second line of equation (48) is obtained from the sum rule

$$\sum_{\mathbf{s}'} q_{\mathbf{s}\mathbf{s}'} = \sum_{\mathbf{s}'} q_{\mathbf{s}'\mathbf{s}} = \lambda_s. \tag{49}$$

The three-dimensional diffusion equations (1) and (2) are obtained from equation (48) by expanding the angle dependence of $I(\mathbf{r},\mathbf{s})$ and $q(\mathbf{s}, sh') \approx q(\mathbf{s}\cdot\mathbf{s}') = q(\cos\gamma)$ in terms of Legendre polynomials and stop after the 1st term (P1 approximation) and then integrating $\mathbf{s}$ over the total solid angle [9, 24].

The two terms of the three-dimensional $I(\mathbf{r},\mathbf{s})$ in P1 approximation are [9, 19]:

$$I(\mathbf{r},\mathbf{s}) = A_{3D} U(\mathbf{r}) + B_{3D}\mathbf{s}\cdot\mathbf{j}(\mathbf{r}) \tag{50}$$

with

$$U(\mathbf{r}) = \sum_{\mathbf{s}} I(\mathbf{r},\mathbf{s})\,, \qquad \mathbf{j}(\mathbf{r}) = \sum_{\mathbf{s}} \mathbf{s} I(\mathbf{r},\mathbf{s})\,,$$

$$A_{3D} = \frac{1}{\sum_{\mathbf{s}}} = 1/4\pi, \qquad B_{3D} = \frac{1}{\sum_{\mathbf{s}} \mathbf{s}\cdot\mathbf{s}} = 3/4\pi.$$

The corresponding expression in one dimension is

$$I(x,\mathbf{s}) = A_{1D}U(x) + B_{1D}\mathbf{s}\cdot\mathbf{j}(x), \tag{51}$$

with $A_{1D} = B_{1D} = 1/\sum_{\mathbf{s}} = 1/2$, which is just equation (10). Since we have shown in the beginning that the diffusion equations (9) are equivalent to the Kubelka-Munk equations (11) we conclude that the P1 approximation, and hence the diffusion approximation in one dimension is exact. This has already been pointed out in [19, 27].

Thus, we can state that the Kubelka-Munk equations (11) are (*i*) identical to the three-dimensional diffusion equation, averaged over the lateral dimensions, and (*ii*) are the proper radiative-transfer equations for the one-dimensional diffuse-reflection problem.

5. Conclusion

We have shown that the Kubelka-Munk equations are identical to the one-dimensional diffusion equation, which is obtained by averaging the three-dimensional diffusion equation with respect to the lateral directions. We obtain as Kubelka-Munk parameters $K = \lambda_a$ (absorptive inverse scattering length) and $S = \frac{3}{2}\lambda_t = \frac{3}{2}\lambda_s(1 - \langle\cos\gamma\rangle)$, where λ_t and λ_s are the transport and scattering inverse scattering lengths, and $\langle\cos\gamma\rangle$ is the anisotropy parameter. Using the 1D diffusion equation we have derived the formulae for a diffuse reflection and transmission, which includes possible internal reflections. In the absence of internal reflections these expressions reduce to those given by Kubelka and Munk. We have demonstrated that the Kubelka-Munk equations are the appropriate radiative transfer equations for the reflection problem with plane-wave incidence (uniform illumination).

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Appendix. Diffusion and the telegrapher's equation

If we include the time dependence, the P1-approximated radiative-transfer equations do not give a diffusion equation, but instead a telegrapher's equation [19]. This equation preserves the $\lambda_a - \tau$ scaling of the solution $U(\mathbf{r},\tau)$

$$U(\mathbf{r},\tau) = \mathrm{e}^{-\lambda_a\tau}U_0(\mathbf{r},\tau), \tag{A.1}$$

where $U_0(\mathbf{r},\tau)$ is the solution in the absence of absorption.

Durian [20] showed that the most general form of a telegrapher's equation, which preserves this scaling is

$$3\alpha\frac{\partial^2}{\partial\tau^2}U(\mathbf{r},\tau) + 3\big(\lambda_t + \alpha\lambda_a\big)\frac{\partial}{\partial\tau}U(\mathbf{r},\tau) + 3\lambda_a\big(\lambda_t + \alpha\lambda_a\big)U(\mathbf{r},\tau) = \nabla^2U(\mathbf{r},\tau). \tag{A.2}$$

In the steady state, the usual steady-state diffusion equation (with still unspecified prefactor α of λ_a) is obtained. It can be easily checked that equation (A.2) fulfils the $\lambda_a - \tau$ scaling for any value of α.

Durian [20] now argues that for small times, which describes the initial spreading of a point source, the proper wave equation

$$\frac{\partial^2}{\partial \tau^2} U(\mathbf{r}, \tau) = \nabla^2 U(\mathbf{r}, \tau) \tag{A.3}$$

must be recovered. This enforces the value of $\alpha = \frac{1}{3}$, and hence a diffusivity of the form of equation (3).

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Дифузія світла в мутних середовищах з внутрішніми відбиваннями

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Встановлено співвідношення між рівнянням Кубелки-Мунка для опису інтенсивності поширення світла в мутних середовищах та рівнянням одновимірної дифузії, яке отримано шляхом усереднення тривимірного рівняння дифузії за поперечними напрямками. Це дає нам можливість однозначно знайти параметри Кубелки-Мунка та вивести вирази для коефіцієнтів дифузійного відбивання та пропускання з врахуванням впливу внутрішніх відбивань. За відсутності внутрішніх відбивань отримуються формули Кубелки-Мунка для цих коефіцієнтів. Показано, шо співвідношення Кубелки-Мунка є властивими рівняннями випромінювального переносу для задачі про одновимірну дифузію.

Ключові слова: *дифузія, мутні середовища, рівняння переносу*