Applied problems and computational methods in radiative transfer

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Abstract

Light scattering in turbid media is essential for such diverse applications as paper and print, computer rendering, optical tomography, astrophysics and remote sensing. This thesis investigates angular variations of light reflected from plane-parallel turbid media using both mathematical models and reflectance measurements, deals with several applications and proposes novel computational methods for solving the governing equations.

Angular variations of light reflected from plane-parallel turbid media is studied using both mathematical models and reflectance measurements. It is found that the light is reflected anisotropically from all media encountered in practice, and that the angular variations depend on the medium absorption and transmittance and on the angular distribution of the incident light. If near-surface bulk scattering dominates, as in strongly absorbing or highly transmitting media or obliquely illuminated media, relatively more light is reflected in large polar (grazing) angles. These results are confirmed by measurements using a set of paper samples. The only situation with isotropic reflectance is when a non-transmitting, non-absorbing medium is illuminated diffusely, and it is shown that this is the only situation where the widely used Kubelka-Munk model is exactly valid.

A number of applied problems is studied, including reflectance measurements, angle resolved color and point spreading. It is seen that differences in instrument detection and illumination geometry can result in measurement differences. The differences are small and if other sources of error — such as fluorescence and gloss — are not eliminated, the differences related to instrument geometry become difficult to discern. Furthermore, the dependence of point spreading in turbid media on the medium parameters is studied. The asymmetry factor is varied while maintaining constant the optical response in a standardized measurement geometry. It is seen that the point spreading increases as forward scattering becomes more dominant, and that the effect is larger if the medium is low-absorbing with large mean free path. It is argued that the directional inhomogeneity of the scattering medium must be captured by the model to reproduce experimental results. Finally, the angle resolved color of a set of paper samples is assessed both theoretically and experimentally. The chroma decreases and the lightness increases as the observation polar angle increases. The observed differences are clearly large, and a modification of the $L^*a^*b^*$ formalism including angle dependent chromatic adaptation is suggested here to handle this situation.

The long standing issue of parameter dependence in the Kubelka-Munk model is partially explained by recognizing that light reflected from paper samples in standardized measurements has angular variations, and by using the appropriate model in the calculation of the scattering and absorption coefficients.

The radiative transfer (RT) equation is solved with a recently proposed particle method (DFPM), both in standard cases and in cases previously considered intractable. Fluorescence is added to the RT equation, thus including wavelength as an additional dimension, and this equation is solved using DFPM. The discrete RT equation can be written as a system of linear equations, and a comprehensive analysis of the convergence properties of DFPM when solving this type of problems is presented.

Sammanfattning

Ljusspridning är viktigt inom så vitt skilda områden som papper och tryck, datorrendering, tomografi, astrofysik och fjärranalys. Denna avhandling behandlar vinkelmässiga variationer hos ljus som reflekteras från planparallella medier, med hjälp av både mätningar och matematiska modeller, samt undersöker flera tillämpningar och föreslår nya beräkningsmetoder för att lösa ingående ekvationer.

Vinkelmässiga variationer hos ljus som reflekteras från planparallella medier studeras med både matematiska modeller och reflektansmätningar. Det visas att ljus reflekteras anisotropiskt från alla förekommande medier, och att de vinkelmässiga variationerna beror på mediets absorption och transmittans och på vinkelfördelningen hos det infallande ljuset. Om ytnära bulkspridning dominerar, som i kraftigt absorberande eller transmitterande medier och i medier med snett infallande ljus, så ökar den relativa mängden ljus som reflekteras i stora polära vinklar (närmare mediets yta). Dessa resultat bekräftas med reflektansmätningar på ett antal pappersprover. Det enda fallet då ljus reflekteras isotropiskt är då ett icke-transmitterande, icke-absorberande medium belyses diffust, och det visas att detta är det enda fallet då den kända Kubelka-Munk modellen är exakt giltig.

Ett antal tillämpade problem studeras, inkluderande reflektansmätningar, vinkelupplöst färg och punktspridning. Det visas att skillnader i instruments detektor- och belysningsgeometri kan ge upphov till mätskillnader. Dessa skillnader är små och om andra felkällor — som fluorescens och glans — inte elimineras, så blir de geometrirelaterade skillnaderna svåra att urskilja. Vidare så studeras hur punktspridning beror på det spridande och absorberande mediets parametrar. Asymmetrifaktorn varieras medan uppmätt mätvärde i en standardiserad mätgeometri hålls konstant. Man ser att punktspridningen ökar när mediet är mer framåtspridande, och att effekten är större om mediet är lågabsorberande med stor medelfri väg. Det föreslås att den modell som används måste beskriva de laterala skillnader som kan finnas i mediet för att stämma med experiment. Slutligen så undersöks den vinkelupplösta färgen hos en mängd pappersprover både teoretiskt och experimentellt. Färgmättnaden minskar och ljusheten ökar när betraktningsvinkeln ökar och närmar sig mediets yta. De iakttagna skillnaderna är uppenbart stora, och en förändring av $L^*a^*b^*$-formalismen som inbegriper vinkelberoende kromatisk adaption föreslås för att hantera denna situation.

Den sedan länge diskuterade frågan om parameterberoende i Kubleka-Munk-modellen förklaras delvis genom att ta hänsyn till att ljus som reflekteras från papper i standardiserade mätningar är anisotropt, och genom att använda tillämpliga modeller för att beskriva detta när mediets spridnings- och absorptionskoefficienter beräknas.

Strålningstransportekvationen löses med en ny partikelmetod (DFPM), både i standardfall och i fall som andra metoder inte kan hantera. Fluorescens inkluderas i ekvationen som löses med DFPM. Den diskretiserade strålningstransportekvationen kan skrivas som ett system av linjära ekvationer, och en omfattande analys av DFPMs konvergensegenskaper för denna typ av problem presenteras.

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Contents

List of papers

This thesis consists of the following papers, herein referred to by their respective Roman numerals:

- **I** M. Neuman and P. Edström, "Anisotropic reflectance from turbid media. I. Theory," J. Opt. Soc. Am. A **27**, 1032–1039 (2010). *Selected for publication in Virtual Journal for Biomedical Optics, Vol. 5, Iss. 9.*

- **II** M. Neuman and P. Edström, "Anisotropic reflectance from turbid media. II. Measurements," J. Opt. Soc. Am. A **27**, 1040–1045 (2010). *Selected for publication in Virtual Journal for Biomedical Optics, Vol. 5, Iss. 9.*

- **III** P. Edström, M. Neuman, S. Avramidis, and M. Andersson, "Geometry related inter-instrument differences in spectrophotometric measurements," Nord. Pulp Pap. Res. J. **25**, 221–232 (2010).

- **IV** M. Neuman, L. G. Coppel, and P. Edström, "Point spreading in turbid media with anisotropic single scattering," Opt. Express **19**, 1915–1920 (2011). *Selected for publication in Virtual Journal for Biomedical Optics, Vol. 6, Iss. 2.*

- **V** L. G. Coppel, M. Neuman, and P. Edström, "Lateral light scattering in paper - MTF simulation and measurement," Opt. Express **19**, 25181-25187 (2011). *Selected for publication in Virtual Journal for Biomedical Optics, Vol. 7, Iss. 2.*

- **VI** M. Neuman, L. G. Coppel, and P. Edström, "Angle resolved color of bulk scattering media," Appl. Optics **50**, 6555-6563 (2011). *Selected for publication in Virtual Journal for Biomedical Optics, Vol. 7, Iss. 2.*

- **VII** M. Neuman, L. G. Coppel, and P. Edström, "A partial explanation of the dependence between light scattering and light absorption in the Kubelka-Munk model," Nord. Pulp Pap. Res. J. **27**, 426-430 (2012).

- **VIII** M. Neuman, S. Edvardsson, and P. Edström, "A particle approach to the radiative transfer equation with fluorescence," submitted to Opt. Lett.

- **IX** S. Edvardsson, M. Neuman, P. Edström, M. Gulliksson, and H. Olin, "Solving linear equations with the dynamical functional particle method," manuscript under preparation.

Related papers not included in the thesis

N. Johansson, M. Neuman, P. Edström, and M. Andersson, "Separation of surface and bulk reflectance by absorption of bulk scattered light," Appl. Opt. **52**, 4749-4754 (2013).

M. Namedanian, L. G. Coppel, M. Neuman, S. Gooran, P. Edström, and P. Kolseth, "Analysis of optical and physical dot gain by microscale image histogram and modulation transfer functions," accepted for publication in J. Imaging Sci. Techn.

T. Linder, T. Löfqvist, L. G. Coppel, M. Neuman, and P. Edström, "Lateral light scattering in fibrous media," Opt. Express **21**, 7835-7840 (2013).

L. G. Coppel, M. Andersson, M. Neuman, and P. Edström, "Fluorescence model for multi-layer papers using conventional spectrophotometers," Nord. Pulp Pap. Res. J. **27**, 418-425 (2012).

H. Hägglund, O. Norberg, M. Neuman, and P. Edström, "Dependence between paper properties and spectral optical response of uncoated paper," Nord. Pulp Pap. Res. J. **27**, 440-444 (2012).

L. G. Coppel, M. Neuman, and P. Edström, "Extension of the Stokes equation for layered constructions to fluorescent turbid media," J. Opt. Soc. Am. A **29**, 574-578 (2012). *Selected for publication in Virtual Journal for Biomedical Optics, Vol. 7, Iss. 6.*

M. Neuman, P. Edström, M. Andersson, L. Coppel, and O. Norberg, "Angular variations of color in turbid media - the influence of bulk scattering on goniochromism in paper," in *Proc. of 5th European Conference on Colour in Graphics, Imaging and Vision*, (Joensuu, Finland, 2010), pp. 407–413.

1 Introduction

The interaction between light and matter determines how we perceive an object. The reflected light gives rise to our visual sensation but can also give important information about the light scattering properties of the medium by observing such things as spectral reflectance factor, transmittance and opacity, and can also be used to derive color sensations using color appearance models. This thesis is about the assessment and interpretation of light reflectance and quantities derived from it, using both measurements and mathematical models.

Understanding how light interacts with matter and the connection to the resulting optical response is important in various industrial sectors. In the paper and print industries light scattering simulations and measurements are used when developing new products, for quality control in production, and in data exchange [1–3]. Furthermore, light scattering is important in the paint industry [4–6], textile industry [7, 8], food industry [9], and is the basis for computer rendering [10] and medical applications such as optical tomography [11, 12]. Propagation of light in turbid media is also studied in research fields such as atmospheric physics [13] and astrophysics [14], and related problems are studied in, e.g., neutron transport [15] and remote sensing [16]. The results presented in this thesis are generic in the sense that they can be useful in all applications dealing with light scattering.

1.1 Purpose of the thesis

The general purpose of this thesis is to use the possibilities of angle resolved radiative transfer models to better understand angular variations of light reflected from plane-parallel turbid media, with a focus on applications relevant for the paper industry. This will make clear how the angular variations depend on the medium parameters and shed new light on the relation between general radiative transfer theory and simplified models used in industrial applications. Furthermore, the consequences of anisotropic reflectance for several applications will be studied. These include reflectance measurements, angle resolved color and point spreading. This thesis also explores novel computational methods for solving the radiative transfer equation to further increase the applicability of radiative transfer theory.

2 Models of radiative transfer

The most complete description of the interaction between light and matter is quantum electrodynamics, which describes all optical phenomena using probability amplitudes and the peculiar rules for superposition of probability amplitudes in quantum mechanics. This leads to mindboggling insights about such things as lenses and diffraction phenomena, but also to an overly complex formalism [17, 18].

Ignoring the quantum nature of light, Maxwell's equations is an adequate description that expresses the connection between light and electromagnetism. Being a vector field theory, Maxwell's equations encompass the wave nature of light including phase and polarization. Simplifying further and ignoring the wavy nature of light we end up with ray optics, or geometric optics, where light travels in straight lines and bends and bounces according to Snell's law and Fresnel's equations. Adding to this the phenomena of absorption and scattering of these light rays we arrive at what is commonly denoted radiative transfer (RT) theory, by which we mean light propagation in turbid media. It should be mentioned though that RT theory can in some formulations also deal with the polarization of light [19].

We can start developing a model for radiative transfer by considering light of intensity i traveling a distance $\mathrm{d}x$ in a turbid medium. This light can be scattered or absorbed in proportion to corresponding material parameters s and k, leading to a change $\mathrm{d}i$ in light intensity that we can express as $\mathrm{d}i = -(s+k)i\mathrm{d}x$. Now let us assume that light is scattered into another direction, in which we denote the light intensity j. Similarly we have that light in that direction is scattered into the direction where light of intensity i travels. The changes in light intensities i and j can then be written $\mathrm{d}i = -(s+k)i\mathrm{d}x + sj\mathrm{d}x$ and $\mathrm{d}j = -(s+k)j\mathrm{d}x + si\mathrm{d}x$ respectively, adding the contribution from the opposite direction. If traveled distance $\mathrm{d}x$ in the two directions has opposite signs, which is the case if we consider a slab geometry where x corresponds to the depth, and if we divide by $\mathrm{d}x$ we end up with

$$\begin{cases} -\dfrac{\mathrm{d}i}{\mathrm{d}x} = -(s+k)i + sj \\ \dfrac{\mathrm{d}j}{\mathrm{d}x} = -(s+k)j + si, \end{cases} \tag{1}$$

which is the widely used Kubelka-Munk (KM) model [20–22]. As is obvious from this derivation the KM model does not handle angular variations in the light intensity and it is difficult to imagine a situation where light behaves in the way postulated by the KM model. The KM model is used extensively in the paper industry together with standardized d/0 measurements [23] to determine the KM scattering and absorption coefficients (s and k), which is also a standardized procedure [24]. Two different backgrounds, i.e. boundary conditions, are then used at the medium bottom boundary. The paper samples are illuminated diffusely and the detector is located at zero degrees, i.e. in the normal direction of the paper. The medium parameters s and k are used for example to monitor and evaluate steps in the paper manufacturing process, such as beating, bleaching and dyeing, and for prediction of optical properties.

Several limitations of the KM model have been reported, such as the dependence between the scattering and absorption coefficients [25–29], and several explanations have been proposed [30–34], but no one has attacked the fundamental limitations of the KM model. Angular variations of reflected light has been observed or discussed by other authors in connection with the KM model [34–37], but not thoroughly or systematically studied.

If we want the model to describe light scattering into an arbitrary direction we have to sum over all directions, thus introducing an integral term. The intensity i then depends on position x, polar angle θ and azimuthal angle ϕ, and a change in intensity can be written

$$\mathrm{d}i = -(s+k)i\mathrm{d}x + \mathrm{d}x\frac{s}{4\pi}\int_{4\pi} i\mathrm{d}\omega,$$

where we integrate over solid angle ω. Dividing by $\mathrm{d}x$ and using conventional notation we get

$$\frac{\mathrm{d}I}{\mathrm{d}x} = -(\sigma_s+\sigma_a)I + \frac{\sigma_s}{4\pi}\int_{4\pi} I\mathrm{d}\omega,$$

which is the RT equation. Here we have introduced σ_s and σ_a to denote the scattering and absorption coefficients. If we consider a slab geometry the RT equation can be written

$$u\frac{\mathrm{d}I}{\mathrm{d}z} = -(\sigma_s+\sigma_a)I + \frac{\sigma_s}{4\pi}\int_{4\pi} I\mathrm{d}\omega,$$

where z is the depth in the medium and $\mathrm{d}z/u$ (with $u = \cos\theta$) is the projection of the traveled distance on the z-axis. Introducing a weighting function $p(\omega, \omega')$ as kernel in the integral, the equation describes anisotropic single scattering. We then get

$$u\frac{\mathrm{d}I}{\mathrm{d}z} = -(\sigma_s+\sigma_a)I + \frac{\sigma_s}{4\pi}\int_{4\pi} p(\omega,\omega')I\mathrm{d}\omega'. \tag{2}$$

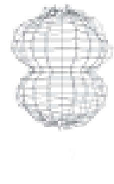
RT theory as expressed in Eq. (2) can be traced back to Lommel [38] via Chwolson [39], Schuster [40] and Chandrasekhar [14] among others.

A commonly used weighting function $p(\omega, \omega')$, or phase function as it is normally called, is the Henyey–Greenstein phase function [41]. It can be written

$$p(\cos\Theta) = \frac{1-g^2}{(1+g^2-2g\cos\Theta)^{3/2}}, \tag{3}$$

where Θ is the angle between the directions of the incident and scattered light. Hence, this phase function does not depend on absolute directions in the medium but only on the direction Θ between incident and scattered light. It contains one parameter, the asymmetry factor g, ranging from -1 to 1 with $g = -1$ meaning complete back scattering, $g = 0$ isotropic scattering and $g = 1$ complete forward scattering. For organic materials such as paper it has been shown that g has values in the interval 0.6–0.9 approximately [42–44], which means that forward scattering dominates.

While it is straightforward to solve the Kubelka-Munk equations (1), the RT equation (2) cannot be solved analytically. The RT equation (2) can instead be solved using e.g. DORT-methods (Discrete Ordinate Radiative Transfer) which is commonplace in many applications [13, 45]. Edström implemented a DORT solution of Eq. (2) in a software tool called DORT2002 [45], which is used extensively in this thesis. Monte Carlo methods can of course also be used [46, 47], but they come with the drawbacks of long simulation times and noise containing solutions.



2.1 Radiative transfer and Maxwell's equations

Maxwell's equations can be solved for simple geometries, but when there are many scattering objects, i.e. when dealing with multi-particle systems, or turbid media, solving Maxwell's equations becomes intractable and established numerical techniques fail. Therefore, one has to resort to approximate solution methods. Radiative transfer (RT) theory is one such approximate solution method.

The relation between radiative transfer theory and Maxwell's equations has been unclear until recently, when Mishchenko derived the RT equation from Maxwell's equations [19, 48–52]. Mishchenko's analysis reveals the necessary conditions to be fulfilled when applying RT theory. These conditions are summarized briefly here and the reader is referred to Mishchenko's work for details.

To go from Maxwells's equations to the RT equation (2) we have to make a series of assumptions. These assumptions are:

1. Non-magnetic material.
2. Far-field scattering – scattering particles and distances between the particles are much larger than the wavelength.
3. Twersky approximation – neglect scattering paths passing the same particle more than once.
4. Ergodicity – averaging over time is equivalent to averaging over state and position.
5. The number of particles is large.
6. Ladder approximation – scattering paths can have no more than one particle in common.
7. Ignore polarization.

When dealing with light scattering in paper, assumptions 1, 4 and 5 are obviously fulfilled. Assumptions 3 and 6 are noncontroversial in practice since their effect can be compensated for by the scattering coefficient.

Assumption 2 is reasonable for light scattering in paper. Components of several microns dominate the paper, and the mean free path is also in the order of several microns. Possible effects such as diffraction and interference are probably negligible since coherent light sources are normally not used in paper applications.

Assumption 7 is made throughout this thesis since we never use polarized illumination or detect the polarization of light. The paper surface is randomized and heterogeneous and polarizes the light only to a small extent. Work related to this is presented by the author in another context [53].

As mentioned previously, the KM model is widely used in industrial applications. For the KM model to be equivalent to the RT equation we have to make further assumptions, as shown in Paper I in this thesis. These assumptions are:

8. Non-absorbing medium.
9. Non-transmitting medium.
10. Isotropic single scattering.
11. Diffuse illumination.

It is obvious that the necessary assumptions can never be fulfilled in practice. KM theory is therefore inherently phenomenological while RT theory is not, since the latter can be related to fundamental principles.

2.2 Fluorescence

Fluorescence is the phenomenon where light of a certain wavelength is absorbed by a fluorophore whereby the fluorophore immediately emits light of a longer wavelength. It is common in the paper industry to add this type of fluorophores (then called fluorescent whitening agents - FWAs) to paper substrates in order to increase perceived whiteness. Ultraviolet light is then absorbed and light is emitted in the blue region of the visible spectrum. A considerable amount of work has been done to extend the Kubelka-Munk model to incorporate also fluorescence [55–59]. These approaches are useful in many situations [60–62], but are stuck with the same fundamental limitations as the standard Kubelka-Munk model outlined in the previous section. In the field of biomedical optics, a modified version of the RT equation is used to simulate fluorescence phenomena [12], where the RT equation then relates a single excitation wavelength to a single emission wavelength. More generally, we have an excitation band and an emission band that can possibly overlap, as is the case with FWAs in paper [60], which gives rise to so-called fluorescence cascades where light is absorbed and emitted in several steps.

The RT equation with fluorescence can for a plane-parallel medium be written

$$\begin{aligned} u\frac{\mathrm{d}I(\lambda)}{\mathrm{d}z} &= -\left[\sigma_s(\lambda)+\sigma_a(\lambda)+\sigma_f(\lambda)\right]I(\lambda) \\ &\quad +\frac{\sigma_s(\lambda)}{4\pi}\int_{4\pi} p(\cos\Theta)I(\lambda)\mathrm{d}\omega \\ &\quad +\frac{1}{4\pi}\int_{\lambda'<\lambda}\int_{4\pi}\sigma_f(\lambda')Q(\lambda',\lambda)I(\lambda')\mathrm{d}\omega\mathrm{d}\lambda', \end{aligned} \tag{4}$$

where we have stated the wavelength dependence of the intensity and parameters explicitly. We have introduced the absorption coefficient σ_f and the quantum yield $Q(\lambda', \lambda)$ of the fluorophore. The quantum yield is the efficiency of transitions from λ' to λ or, equivalently, the ratio between the amount of light absorbed at wavelength λ' and the amount of light emitted at wavelength λ. The third term on the RHS of Eq. (4) includes an integral over wavelength, thus summing all contributions through fluorescence from wavelengths λ' shorter than λ. There is no restriction on the emission and excitation bands, so the equation can describe fluorescence cascades. Equation (4) is considerably more difficult to solve than the RT equation without fluorescence

(2). Monte Carlo methods is therefore a viable option [47, 63, 64], but they of course come with the drawbacks of long simulation times and noise. If we try using DORT methods to solve Eq. (4), after the common steps in the solution procedure we end up with the equation system

$$\begin{cases} \mu_i \dfrac{\mathrm{d}I^{m+}(\tau,\mu_i,\lambda_k)}{\mathrm{d}\tau} = I^{m+}(\tau,\mu_i,\lambda_k) - \dfrac{a}{2}\displaystyle\sum_{j=1}^{N} w_j p^m(\mu_j,\mu_i,\lambda_k) I^{m+}(\tau,\mu_j,\lambda_k) \\ \qquad - \dfrac{a}{2}\displaystyle\sum_{j=1}^{N} w_j p^m(-\mu_j,\mu_i,\lambda_k) I^{m-}(\tau,\mu_j,\lambda_k) - X_{0i}^{m+}\exp(-\tau/\mu_0) \\ \qquad - \delta_{m0}\dfrac{1}{4\pi}\displaystyle\sum_{l=1}^{k-1}\sum_{j=1}^{N}\sum_{q=1}^{P}\sum_{n=0}^{2N-1} w_j \Delta\lambda_l \Delta\varphi_q \bar{a}_f(\lambda_l,\lambda_k) \\ \qquad \cdot Q(\lambda_l,\lambda_k) I^{n+}(\tau,\mu_j,\lambda_l)\cos(n(\varphi_0-\varphi_q)) \\ \qquad - \delta_{m0}\dfrac{1}{4\pi}\displaystyle\sum_{l=1}^{k-1}\sum_{j=1}^{N}\sum_{q=1}^{P}\sum_{n=0}^{2N-1} w_j \Delta\lambda_l \Delta\varphi_q \bar{a}_f(\lambda_l,\lambda_k) \\ \qquad \cdot Q(\lambda_l,\lambda_k) I^{n-}(\tau,\mu_j,\lambda_l)\cos(n(\varphi_0-\varphi_q)) \\ \qquad - \delta_{m0}\dfrac{1}{4\pi}\displaystyle\sum_{l=1}^{k-1} \exp(-\tau_l/\mu_0)\Delta\lambda_l I_{0b}(\lambda_l)\bar{a}_f(\lambda_l,\lambda_k) Q(\lambda_l,\lambda_k) \\ -\mu_i \dfrac{\mathrm{d}I^{m-}(\tau,\mu_i,\lambda_k)}{\mathrm{d}\tau} = I^{m-}(\tau,\mu_i,\lambda_k) - \dfrac{a}{2}\displaystyle\sum_{j=1}^{N} w_j p^m(\mu_j,-\mu_i,\lambda_k) I^{m+}(\tau,\mu_j,\lambda_k) \\ \qquad - \dfrac{a}{2}\displaystyle\sum_{j=1}^{N} w_j p^m(-\mu_j,-\mu_i,\lambda_k) I^{m-}(\tau,\mu_j,\lambda_k) - X_{0i}^{m-}\exp(-\tau/\mu_0) \\ \qquad - \delta_{m0}\dfrac{1}{4\pi}\displaystyle\sum_{l=1}^{k-1}\sum_{j=1}^{N}\sum_{q=1}^{P}\sum_{n=0}^{2N-1} w_j \Delta\lambda_l \Delta\varphi_q \bar{a}_f(\lambda_l,\lambda_k) \\ \qquad \cdot Q(\lambda_l,\lambda_k) I^{n+}(\tau,\mu_j,\lambda_l)\cos(n(\varphi_0-\varphi_q)) \\ \qquad - \delta_{m0}\dfrac{1}{4\pi}\displaystyle\sum_{l=1}^{k-1}\sum_{j=1}^{N}\sum_{q=1}^{P}\sum_{n=0}^{2N-1} w_j \Delta\lambda_l \Delta\varphi_q \bar{a}_f(\lambda_l,\lambda_k) \\ \qquad \cdot Q(\lambda_l,\lambda_k) I^{n-}(\tau,\mu_j,\lambda_l)\cos(n(\varphi_0-\varphi_q)) \\ \qquad - \delta_{m0}\dfrac{1}{4\pi}\displaystyle\sum_{l=1}^{k-1} \exp(-\tau_l/\mu_0)\Delta\lambda_l I_{0b}(\lambda_l)\bar{a}_f(\lambda_l,\lambda_k) Q(\lambda_l,\lambda_k), \end{cases}$$

$$m = 0,\ldots,2N-1, \quad i = 1,2,\ldots,N.$$

for each Fourier component m in the series expansion of the azimuthal dependence (further details omitted). This can be written in vector-matrix form as

$$\frac{\mathrm{d}}{\mathrm{d}\tau}\begin{bmatrix}\mathbf{I}^+ \\ \mathbf{I}^-\end{bmatrix} = \begin{bmatrix}-\alpha & -\beta \\ \beta & \alpha\end{bmatrix}\begin{bmatrix}\mathbf{I}^+ \\ \mathbf{I}^-\end{bmatrix} - \begin{bmatrix}\mathbf{Q}_b^+ \\ \mathbf{Q}_b^-\end{bmatrix} - \begin{bmatrix}\mathbf{Q}_f^+ \\ \mathbf{Q}_f^-\end{bmatrix}. \tag{5}$$

It is straightforward to find the homogeneous solution to Eq. (5). However, the particular solution cannot be found in the same straightforward manner since the ansatz for particular solution has to contain the same dependence on depth in the medium as the last term $\mathbf{Q}_f^{\pm}$. This term is the light fluoresced to the current wavelength from shorter wavelengths, possibly in several steps. It can be calculated numerically at specified depths but we need a differentiable expression to use as ansatz. This can be accomplished by e.g. fitting a series of exponentials to the numerical data, but it is not at all obvious how to construct a generic solution method working for all possible boundary conditions and parameter setups. During the work with this thesis the author therefore started to look for other options.

3 Particle methods

The concept of particle methods is usually associated with physical particle methods, which have been used to solve problems in e.g. celestial mechanics [65, 66]. Edvardsson and Uesaka recently exploited the usefulness of particle methods when simulating the open draw section of a paper machine [67]. A comprehensive review of physical particle methods is provided by Li and Liu [68].

The particles can in this case represent either physical entities or quasi-particles that interact with each other and the surrounding. Having defined the forces present, the movement of each particle is calculated by integrating the equations of motions in time. This type of particle methods is conceptually appealing in many situations, mainly because problems previously considered intractable when describing them with differential equations nowadays can be solved fairly easy on a standard computer. A differential equation (such as the Navier-Stokes equation of fluid dynamics or the wave equation) is often derived by considering infinitesimally small elements and the forces that act on these elements. If the equation contains terms that make the equation difficult to solve, they are often neglected. Furthermore, complex boundary conditions can be difficult to handle, and to obtain a solution the equation is often discretized and solved numerically. In doing this we have passed from one discrete formulation (the infinitesimally small elements) to another (the discretized equation) where the connection to the underlying physics is somewhat unclear. The idea with physical particle methods is to stick with the original particle formulation, as close as possible to the underlying physics.

3.1 The Dynamical Functional Particle Method

Generalizing the concept of particle methods, it was regognized by Edvardsson et al. [69] that similar ideas can be used to solve general functional equations, and the Dynamical Functional Particle Method (DFPM) was developed. In this method the discretized functional equation $\mathcal{F} = 0$ is solved by treating each grid point, or particle, as a harmonic oscillator that evolves in an artificial evolution time. When the oscillators are at rest the functional is zero and the solution is thereby obtained. The crucial point is to make the oscillators critically damped for fast convergence. This is generally not possible and the best compromise has to be found.

Let us consider the RT equation with fluorescence (4) as an example. A discrete version of this equation can be written

$$u_j \frac{I^l_{i+1jk} - I^l_{i+1jk}}{2h_z} = -\sigma^l_e I^l_{ijk} + \frac{\sigma^l_s}{2N_a} \sum_{m=1}^{N_p} \sum_{n=1}^{N_a} p_{mn;ij} I^l_{imn} \omega_m + \frac{1}{2N_a} \sum_{q<l} \sum_{m=1}^{N_p} \sum_{n=1}^{N_a} \sigma^q_f Q_{ql} I^q_{ijk} \omega_m, \tag{6}$$

where indices i, j, k, l correspond to z, θ, ϕ, λ respectively, $\sigma_e = \sigma_s + \sigma_a + \sigma_f$ is the extinction coefficient, N_p and N_a are the number of quadrature points in polar and azimuth angle, and ω is a quadrature weight in polar angle. The functional is defined by writing all terms in Eq. (6) on the RHS;

$$\mathcal{F}^l_{ijk} = -u_j \frac{I^l_{i+1jk} - I^l_{i+1jk}}{2h_z} - \sigma^l_e I^l_{ijk} + \frac{\sigma^l_s}{2N_a} \sum_{m=1}^{N_p} \sum_{n=1}^{N_a} p_{mn;ij} I^l_{imn} \omega_m + \frac{1}{2N_a} \sum_{q<l} \sum_{m=1}^{N_p} \sum_{n=1}^{N_a} \sigma^q_f Q_{ql} I^q_{ijk} \omega_m,$$

and we see that when the functional is zero Eq. (6) is solved. We now introduce a dependence of I on artificial time and formulate DFPM as

$$\mathcal{F}^{lt}_{ijk} = \ddot{I}^{lt}_{ijk} + \mu \dot{I}^{lt}_{ijk}, \tag{7}$$

where index t corresponds to artificial time and $\dot{I}^{lt}_{ijk}$ and $\ddot{I}^{lt}_{ijk}$ are the first and second order derivatives of I^{lt}_{ijk} with respect to artificial time, and where μ is a damping coefficient. We can then see that Eq. (7) is analogous to a harmonic oscillator. Particle $ijkl$ thus oscillates in artificial time and dissipates energy in proportion to the damping coefficient μ. It should be noted here that there is no known restriction on the functional, i.e. the force on the particles. It can for example be nonlinear.

Equation (7) is integrated in artificial time for all particles using a time step Δt to arrive at a solution. It has proved advantageous to use a symplectic integrator [70,71] since it is cost efficient, stable and allows for a large time step. The accuracy is not particularly good, but this is no a disadvantage in this case since the actual path to the solution is not of interest, but merely the solution at convergence. We therefore integrate Eq. (7) as

$$\dot{I}^{l,t+1}_{ijk} = \dot{I}^{lt}_{ijk} + \left(\mathcal{F}^{lt}_{ijk} - \mu \dot{I}^{lt}_{ijk} \right) \Delta t$$
$$I^{l,t+1}_{ijk} = I^{lt}_{ijk} + \dot{I}^{l,t+1}_{ijk} \Delta t.$$

A considerable amount of analysis on how to find optimal damping μ and time step Δt is done in Paper IX in this thesis. Near-optimal parameters can be found empirically by starting with a short time step to find an optimal damping. The time step is then increased until the method becomes unstable and a new optimal damping is then found by trial-and-error. This process can be repeated until there is no further improvement in convergence properties.

An important result from Paper IX for solving the RT equation with DFPM is provided here. Writing Eq. (6) on vector-matrix form ($Ax = b$), a near-optimal damping for time step Δt is given by

$$\mu = \frac{2 - (\mathrm{tr}(A)/n)\Delta t^2}{\Delta t},$$

where n is the number of elements in x and $\mathrm{tr}(A)$ is the trace of A. This expression is useful to find the part of the $(\mu, \Delta t)$-space where the method converges, which can be quite tedious in some cases using a trial-and-error-method. The matrix corresponding to the RT equation has in general complex eigenvalues. If the eigenvalues are real valued there are closed form expressions for both the optimal damping and the time step as given in Paper IX. Further research on the convergence properties of DFPM is currently ongoing.

4 Some comments on terminology and notation

Terminology and notation related to light scattering differ somewhat depending on the application area. To avoid confusion, these matters are clarified in this section.

The intensity I in RT theory is called radiance and denoted L in radiometry [72]. The radiance is defined as

$$L = \frac{\mathrm{d}^2 P}{\cos\theta \mathrm{d}A \mathrm{d}\omega}, \tag{8}$$

where $\mathrm{d}P$ is radiant power, $\mathrm{d}A$ is a surface element, $\mathrm{d}\omega$ is a solid angle and θ is the angle between the normal of $\mathrm{d}A$ and $\mathrm{d}\omega$. The radiance thus has the unit $[\mathrm{W} \cdot \mathrm{m}^{-2} \cdot \mathrm{sr}^{-1}]$ and is the amount of energy per time passing through a surface in a certain direction contained in a solid angle, divided by the product of that solid angle and the projection of the surface area on the direction orthognoal to the direction of propagation.

In applications dealing with light transport in biological tissue the radiance is often denoted ψ (see e.g. Klose and Larsen [73]) and the RT equation is stated as

$$\boldsymbol{\Omega} \cdot \nabla\psi(\mathbf{r}, \boldsymbol{\Omega}) + \mu_t(\mathbf{r})\psi(\mathbf{r}, \boldsymbol{\Omega}) = \mu_s(\mathbf{r}) \int_{2\pi} p(\boldsymbol{\Omega}, \boldsymbol{\Omega}')\psi(\mathbf{r}, \mathbf{r}')\mathrm{d}\Omega'$$

where $\boldsymbol{\Omega}$ is the direction, $\mathbf{r}$ is the position and μ_t and μ_s are the attenuation (extinction) and scattering coefficients respectively, thus adding further to the plethora of notation.

Another commonly used quantity is called flux and denoted F in RT theory. It is called irradiance and denoted E in radiometry. The irradiance at a point is the amount of energy per time incident on an infinitesimal surface containing the point. It has the unit $[\mathrm{W} \cdot \mathrm{m}^{-2}]$ and can be written

$$E = \frac{\mathrm{d}P}{\mathrm{d}A}.$$

The radiant exitance M is the corresponding quantity for emittance, i.e., at a point it is the amount of energy per time emitted by an infinitesimal surface element containing the point.

A convenient and widely used construction for relating the irradiance to the radiance is the bidirectional reflectance-distribution function (BRDF), denoted f_r [74]. Its general definition is

$$\mathrm{d}L(\theta_i, \varphi_i, \theta_r, \varphi_r, E) = f_r(\theta_i, \varphi_i, \theta_r, \varphi_r)\mathrm{d}E(\theta_i, \varphi_i), \tag{9}$$

and we see that it gives the reflected radiance as a function of irradiance and angles of incidence (subscript i) and reflection (subscript r). The BRDF $f_r(\theta_i, \varphi_i, \theta_r, \varphi_r)$ can be obtained by solving the RT equation, and the radiance can be obtained in a specific solid angle by integrating Eq. (9) over this solid angle. This is impossible in practice and a simpler version of the BRDF is often used to represent angle resolved reflectance data given that a certain illumination is used. That is, we are not interested in a BRDF containing information about how changing the illumination affects the reflected light. This would then correspond to another measurement or simulation setup. We can therefore remove the dependence on θ_i, φ_i in the BRDF and integrate Eq. (9) over θ_i, φ_i containing all incident light. Furthermore, if we assume that all measurements are done in-plane, so that we get no dependence on φ_r (which is equivalent to assuming azimuthal symmetry), Eq. (9) becomes

$$L(\theta_r) = f_r(\theta_r)E. \tag{10}$$

It follows from Mishchenko's work [19,48–52] that the radiance can be measured with a well-collimated radiometer, and we can therefore assess also the BRDF experimentally through Eq. (10) by careful calibration of the measurement instrument.

5 Summary of the papers and contribution of the thesis

Papers I-VII included in this thesis all use radiative transfer models to investigate various aspects of angle resolved light scattering. The main contribution of Papers I-VII is new knowledge about the interaction of light with plain-parallel turbid media through the application of these models. This is relevant for virtually any area where light scattering is an important part. The application areas in this thesis include reflectance measurements, angle resolved color and point spreading. Paper VIII exploits the recently proposed Dynamical Functional Particle Method (DFPM) for solving the radiative transfer equation in standard cases but also in cases previously considered intractable. Paper IX is different in the sense that it is concerned exclusively with numerical linear algebra, solving systems of linear equations with DFPM. Results in this paper are necessary to attack the applied problems in Paper VIII, but it also contributes extensively to the field of numerical linear algebra.

5.1 Paper I

Anisotropic reflectance from turbid media. I. Theory

Paper I employs the DORT2002 radiative transfer model to show that the light reflected from turbid media is anisotropic in all cases encountered in practice and that the anisotropy depends on the medium properties in a characteristic way. This is illustrated by displaying the simulated BRDF of three media. These show that the BRDF is close to isotropic when the medium is low-absorbing and opaque, while it is anisotropic when there is considerable absorption or transmittance. To understand this, simple expressions are stated showing that the angular variations of the reflectance depend on the relative contribution from different scattering depths in the medium. When near-surface bulk scattering increases, the amount of light reflected in large polar angles increases. This is the case for optically thin or strongly absorbing media, or when the illumination is incident obliquely. The illumination dependence is predicted by theory and verified by simulation in this paper. These results also lead to the conclusion that there exists no bulk scattering ideal diffusor. If the light scattering properties are ideal it will still reflect anisotropically depending on the illumination. It reflects isotropically only when illuminated diffusely. This has consequences for the wide spread Kubelka-Munk model. It is shown that the Kubelka-Munk model is exactly valid only under very specific conditions. These are zero transmittance, zero absorption and diffuse illumination. Neglecting the angular variations in the reflected light introduces errors that can be 20-40 %, depending on the medium parameters. It is also pointed out that the error introduced when applying the Kubelka-Munk model can be minimized. For example, by placing the detector in an optimal angle in a measurement setup.

Paper I was written in cooperation with Prof. Per Edström. The contribution of the author of this thesis was to perform simulations, to do calculations and main part of the analysis and writing.

5.2 Paper II

Anisotropic reflectance from turbid media. II. Measurements

Paper II confirms experimentally the theoretical results from Paper I. The angle resolved reflectance from a set of paper samples varying in dye and filler content and thickness is measured using a goniophotometer. The addition of dye and fillers alters the scattering and absorbing properties of the samples so that their influence on the angle resolved reflectance can be studied. Corresponding simulations are also done using DORT2002 and it is seen that the agreement between measurements and simulations is good. Furthermore, the angle resolved reflectance of a $BaSO_4$ diffusor is measured and simulated with varying angle of incidence of the illumination. It is seen that the agreement between experiment and theory is good, and that the diffusor is not ideal. Paper II also discusses how anisotropy of the reflected light can affect standardized measurements through e.g. the calibration routine which often involves alleged ideal diffusors.

Paper II was written in cooperation with Prof. Per Edström. The contribution of the author of this thesis was to do measurements, simulations, and main part of the analysis and writing.

5.3 Paper III

Geometry related inter-instrument differences in spectrophotometric measurements

Paper III investigates differences in measured reflectance factor between the 45/0 and d/0 instrument geometries. The effect of external factors, such as sample background, calibration and sample inhomogeneity, on the reflectance measurements is quantified and a method for eliminating them is suggested. The remaining instrument differences can be attributed to the instrument geometry. The 45/0 and d/0 reflectance factors are measured and compared for a set of paper samples. The instruments are also simulated using DORT2002 and it is seen that the inter-instrument differences are of the same magnitude as in the measurements. The inter-instrument differences are small and around 0.1 ΔE^*_{ab} for the set of paper samples. An explanation is proposed for the geometry induced inter-instrument differences where results from Papers I and II are used. The mean scattering depth in the 45/0 instrument is larger than in the d/0 instrument. This means that the light reflected from a medium with the respective illuminations will differ in angular dependence. Relatively more light will be reflected towards the detector with the 45/0 illumination if the medium is low absorbing, highly scattering and opaque. The d/0 instrument will on the other hand detect a higher reflectance if the medium is strongly absorbing or transmitting, since then more light is reflected with the d/0 illuminations than with the 45/0 illumination.

Paper III was written in cooperation with Prof. Per Edström, M.Sc. Stefanos Avramidis and Dr. Mattias Andersson. The contribution of the author of this thesis was to do part of the writing and analysis behind the proposed explanation.

5.4 Paper IV

Point spreading in turbid media with anisotropic single scattering

Paper IV concerns point spreading in turbid media. The same set of dyed paper samples as in Paper II and VI is used to assess relevant medium properties describing the scattering characteristics. The d/0 reflectance factor is measured and the medium properties albedo and mean free path are calculated using DORT2002 for a set of values of the asymmetry factor. In this way the optical response in the d/0 instrument will be the same irrespective of the value of the asymmetry factor. The point spreading is then simulated for all media using the Monte Carlo model Open PaperOpt [46]. A numerical measure is introduced to quantify point spreading. It is seen that the point spreading measure increases linearly with the asymmetry factor for both opaque and thin media. It is largest for opaque media with high albedo and large mean free path. The number of scattering events that the wave packets undergo before leaving the medium is also calculated using the Monte Carlo model. Increasing the asymmetry factor increases the number of scattering events. Media with high albedo have a similar contribution to the reflectance from different scattering orders. Media with low albedo are also similar in this respect. This means that the point spreading is determined by the distance that the multiply scattered light can travel. This distance is larger if the mean free path is large and absorption is low. A medium with high albedo and large mean free path will thus give the largest point spreading, and it will increase further if light is scattered more in the forward direction.

The presented results thus show that the asymmetry factor plays a significant role in point spreading together with the albedo, mean free path and thickness. This means that models of point spreading must take all of these medium characteristics into account in order not to be based on ad hoc assumptions.

Paper IV was written in cooperation with Dr. Ludovic Coppel and Prof. Per Edström. The contribution of the author of this thesis was to perform simulations and measurements, to do calculations and main part of the analysis and writing.

5.5 Paper V

Lateral light scattering in paper — MTF simulation and measurement

Paper V continues the line of research in Paper IV by comparing Monte Carlo simulations of the modulation transfer function (MTF) of 22 paper samples with the corresponding measurements. The simulations show that the asymmetry factor has a considerable impact on the MTF, as is expected from the results in Paper V. The inverse frequency at half maximum of the MTF (denoted k_p) is commonly used as a single metric of the MTF. Paper V shows that k_p is an inappropriate metric since it does not reflect the variations in MTF as the asymmetry factor is varied or the differences between the Arney model and the Monte Carlo simulations.

The Monte Carlo simulations cannot explain the large lateral scattering observed in the measurements. It is argued that this is due to the directional inhomogeneity in the paper samples, and that models aiming at describing lateral scattering in paper and other fibrous structures must include scattering parameters that depend on absolute directions.

Paper V was written in cooperation with Dr. Ludovic Coppel and Prof. Per Edström. The contribution of the author of this thesis was to do part of the analysis and writing.

5.6 Paper VI

Angle resolved color of bulk scattering media

Paper VI deals with the consequences of the findings in Papers I and II for angle resolved color. The same set of dyed paper samples is used here as in Paper I and II. The angle resolved spectral reflectance factor of the samples is measured with a goniophotometer at InFotonics Center, Joensuu. The medium scattering and absorption coefficients are assessed using d/0 measurements and assuming that $g = 0.8$. In this way the goniophotometer measurements can be simulated using DORT2002 since we then have the necessary parameter setup. The reflectance factor increases as the polar angle increases in both measurements and simulations. Simulations and measurements agree well. The relative increase is larger for wavelengths in the absorption band of the dye. This is in agreement with the results of Papers I and II, and the characteristics of the anisotropy are further confirmed by these result. Note that the samples are prepared in such a way to minimize gloss and that the simulations contain no surface scattering. These effects are therefore a result of bulk scattering, and not of surface scattering. The measured and simulated angle resolved reflectance factor spectra are then converted to the $L^*a^*b^*$ color space using the D50 illuminant and 2° observer. It is seen that the angular variations in reflectance factor give rise to strikingly large variations in the angle resolved $L^*a^*b^*$ values. The maximum CIE 1976 color difference ΔE^*_{ab} is obtained for the heavily dyed paper. It is 23.9 for the measurements and 30.5 for the simulations. This is far above the limit of what is possible to perceive. We therefore suggest angle resolved $L^*a^*b^*$ coordinates and evaluate alternatives to the ideal diffuser as reference white, i.e. we suggest angle dependent chromatic adaptation. The estimated color variations are then reduced, but they are still unrealistically large. It is argued that the human visual system has a limited sensitivity to changes in chroma.

A result of the simulations and the measurements is that the lightness increases and the chroma decreases when the observation polar angle is increased. An explanation is proposed for this based on the findings in Papers I and II. The lightness increases since the light of all wavelengths is reflected anisotropically with more light in large polar angles. The chroma decreases since light of wavelengths that is absorbed by the dye is reflected even more anisotropically from the medium, with a larger relative increase in large polar angles.

Paper VI was written in cooperation with Dr. Ludovic Coppel and Prof. Per Edström. The contribution of the author of this thesis was to perform simulations, to do calculations and main part of the analysis and writing.

5.7 Paper VII

A partial explanation of the dependence between light scattering and light absorption in the Kubelka-Munk model

Paper VII proposes an explanation of the long debated Kubelka-Munk anomaly where the scattering and absorption coefficients appear to be interdependent. It is argued that the anisotropy of the reflected light in combination with the d/0 instrument geometry, which does not detect the anisotropy, is causing the Kubelka-Munk anomaly. This anisotropy is explained and investigated in Papers I and II. Using angle resolved radiative transfer theory, thus taking the anisotropy into account, the interdependence is much less significant. Using the more comprehensive radiative transfer theory, this type of geometry induced error can thus be avoided. A measure is introduced to illustrate where in the (R_0, R_∞)-space the models deviate the most, and it is shown that the deviation is small in low absorbing optically thick media.

Paper VII was written in cooperation with Dr. Ludovic Coppel and Prof. Per Edström. The contribution of the author of this thesis was to perform simulations, to do calculations and main part of the analysis and writing.

5.8 Paper VIII

A particle approach to the radiative transfer equation with fluorescence

Paper VIII uses the recently developed Dynamical Functional Particle Method (DFPM) to solve the RT equation. DFPM gives several advantages compared to traditional solution methods, such as discrete ordinates. It is straightforward to implement and converges rapidly for standard problems. DFPM handles arbitrary boundary conditions, and an example is provided where the incident illumination consists of two beams, which is not handled by standard DORT solution methods. Additionally, DFPM can handle arbitrary phase functions, including those dependent on absolute directions in the medium. DORT methods are numerically unstable when the single scattering is strongly forwardly peaked. This is a well known issue and a considerable amount of work has be done to overcome this. Paper VIII shows that DFPM does not suffer from this problem. Finally, the RTE including fluorescence is solved using DFPM. The excitation and emission bands of the fluorophore are allowed to overlap, thus including the so-called fluorescence cascade. It is argued that DFPM is a promising method to handle even more complex RT problems.

Paper VIII was written in cooperation with Prof. Sverker Edvardsson and Prof. Per Edström. The contribution of the author of this thesis was coding, numerical testing and calculations, and main part of the analysis and writing.

5.9 Paper IX

Solving linear equations with the dynamical functional particle method

Paper IX analyzes in detail the properties of the Dynamical Functional Particle Method when solving systems of linear equations. The method is compared with standard methods such as the conjugate gradient method, Gauss-Seidel and Jacobi iterations. It is shown that the performance of DFPM is superior to all standard methods with the exception of the conjugate gradient method, which is surpassed only for large matrices with a large span in the eigenvalues. However, DFPM handles asymmetric matrices while the conjugate gradient method does not. It is recognized that the computational complexity of DFPM has the potential to scale linearly with the matrix size for sparse matrices, which would revolutionize the field of numerical linear algebra. In order for this to happen all oscillators in DFPM must be critically damped.

Paper IX was written in cooperation with Prof. Sverker Edvardsson, Prof. Per Edström., Prof. Mårten Gulliksson and Prof. Håkan Olin. The contribution of the author of this thesis was some numerical testing, analysis and writing.

6 Discussion

We have seen that the light reflected from turbid media has angular variations in all situations encountered in practice. Angle resolved radiative transfer theory is necessary to describe these variations. Paper I showed that the variations depend on the angular distribution of the incident light and the medium absorption and transmittance. Varying these factors will alter the mean scattering depth in the medium and thereby the angular distribution of the reflected light. Furthermore, we saw in Paper II that the theoretical predictions could be confirmed experimentally through goniophotometric measurements of light reflectance from paper samples.

These findings can potentially have a large impact on many applications dealing with measurements of scattered light. For example, we showed that a non-transmitting, low-absorbing medium, i.e. what is normally thought of as an ideal diffusor, reflects light anisotropically depending on the angular distribution of the illumination. This type of medium, such as a $BaSO_4$ plate, is often used for calibration purposes, and neglecting angular variations can introduce errors. It should be mentioned though, that the error can cancel to some extent in some cases, for example if measurements are done to compare samples. But it is still an uncomfortable situation since this is not controlled.

Paper III showed that this kind of anisotropic light reflectance will result in different instrument readings in instruments with different illumination and detection geometries. Other sources of error in reflectance measurements, such as fluorescence, gloss and different backgrounds, can give far larger differences between instruments. This means that if these other sources of error are not carefully eliminated, the error due to instrument geometry will be difficult to discern. However, if the measurements are perfect in all other respects, the instrument geometry can have a considerable impact on the measurement results.

Paper IV and V discuss point spreading in turbid media, and constitute an example of how results concerning light scattering are important for such diverse fields as halftone image reproduction, light propagation in tissue and computer rendering. We saw that assumptions about the single scattering anisotropy can greatly alter the average lateral distance that incident light travels before exiting the medium, but still give the same optical response in a specific measurement geometry. This illustrates why angle resolved models and measurements are crucial for a full understanding of light scattering in turbid media. It also shows that model simplification can be a risky business since important phenomena can be ignored and since the simplified model can be used by others, unaware of its limitations.

Angle resolved color was investigated both experimentally and theoretically in Paper VI. The results gave rise to several new questions. The measured and simulated differences in color when varying the observation angle were clearly large – larger than what is perceived by the human eye. We therefore proposed an extension of the $L^*a^*b^*$ formalism to handle angle resolved color where the chromatic adaptation is angle dependent. Using the proposed modifications the angular color differences decrease, but it is still an open issue as to what extent the human visual system is less sensitive to changes in chroma, and the experimental verification of the proposed solution remains to be done. This is a fairly complicated matter since it involves perception studies with varying observation angles.

Paper VII adds to the increased knowledge gained when using general RT theory instead of the KM model. The long standing issue of parameter dependence in the KM model could be partially explained by recognizing that the light reflected from the paper samples in the measurement situation is anisotropic. Using general RT we were able to incorporate this anisotropy into the calculations of the scattering and absorption coefficients. Previous extensive discussion about this matter has not recognized this possibility, which is most probably due to the tradition in a particular field and reluctance to adopt methods from other fields.

Paper VIII is a first attempt to exploit the possibilities of the mathematical particle method DFPM. This method is promising for solving even more complex problems than the ones treated here. These can include RT in three spatial dimensions with medium inhomogeneities, irregularly shaped media or phenomena such as phosphorescence and internal sources. Extending the method to handle the full Stokes four-vector, thus including polarization can probably also be done. Paper IX is an important step in understanding the convergence properties of DFPM. As the number of grid points (i.e. particles) increases, the computational complexity of the method becomes crucial. DFPM has already shown promising for handling very large problems such as the ones outlined above, and Paper IX shows that it is superior in some cases to standard methods such as the conjugate gradient method.

7 Future work

The Dynamical Functional Particle Method (DFPM) has shown promising for solving general functional equations and more specifically, in this thesis, systems of linear equations. DFPM should be developed further and refined. There is still more potential to attack larger problems and to solve them faster than established methods. Research on this is currently ongoing.

DFPM is also a viable method for solving optimization problems, and it can be a candidate for solving the parameter estimation problem to determine the spectral asymmetry factor g from goniophotometer measurements.

The concept of particle methods can be developed in light scattering applications by considering the Discrete Dipole Approximation (DDA) [75]. DDA solves Maxwell's equation in arbitrary geometries by making use of a large number of oscillating dipoles. This approach ends up in a large equation system, and it would be interesting to see if DDA could be combined with DFPM to give an efficient solution procedure for arbitrary geometries. Such a tool would be valuable in understanding photonic structures and structural color, often found in animal species [76]. Adding quantum phenomena such as fluorescence to the DDA framework could also prove interesting.

As mentioned above, it is still an open issue how to handle angle resolved color.

Relevant perception studies where the observation angle can be varied could answer many questions and lead to new international agreements on how to handle angle resolved color.

To further increase the usefulness and explanative power of the combination of models and measurements in paper applications more elaborate models of fibrous structures such as paper have to be developed. These must most probably include some sort of surface phenomena. A laterally resolved RT model with directional inhomogeneities combined with a surface model would be a good candidate. Another concept could be to develop the DDA in this direction and to combine it with an efficient numerical solver such as DFPM.

The possibility to solve the RT equation including fluorescence can give new insight into the effect of fluorescence on anisotropy and phenomena related to it, and will open up possibilities to optimize for example the paper composition for increased whiteness.

8 Conclusions

We have seen that the angular variations of light reflected from turbid media depend on the medium parameters in a characteristic way. Radiative transfer models have to be angle resolved to capture this dependence. Simplified models that ignore angular variations, such as the Kubelka–Munk model, are widely used in industry. Applying these models to, e.g., reflectance measurements can introduce errors. The magnitude of the error depends on the particular situation and the medium properties, but it is significant for a large range of parameters. Using angle resolved radiative transfer models relieves this uncomfortable situation and opens up for a better understanding of light reflectance from turbid media. The specific instrument geometry can then be taken into account and the model parameters are physically objective, i.e. they can be given a meaning outside the model. The explanative power of the model–measurement compound is therefore far greater in this case compared to when using simplified models. Furthermore, angle resolved models can give important guidelines when designing new instrument geometries. The instrument geometry can be made optimal in some sense, for example by minimizing the geometry induced error for a given set of medium parameters.

This thesis has focused on light scattering applications in the paper industry, but the results presented here can be important for many applications also outside the paper industry. We saw here that, apart from reflectance measurements, angle resolved color and point spreading is better understood using angle resolved radiative transfer. The color appearance of a light scattering material is of great importance for example in the paper, print and packaging industries. The proposed modification of the $L^*a^*b^*$ formalism suggesting an angle dependent chromatic adaptation could have significant impact on standardized ways of quantifying color. A correct understanding of point spreading is essential, for example, to compensate for optical dot gain in halftone prints and to render realistic computer graphics.

Employing the Dynamical Functional Particle Method for solving the RT equation opens up several new possibilities to attack large and complex problems. These can be scattering in inhomogeneous, irregularly shaped three-dimensional media with internal sources and phosphorescence or fluorescence. Furthermore, DFPM is a general method for solving functional equations, and the results presented here has contributed to its development. Solving systems of linear equations faster than established methods, and also systems previously unsolvable with comparable methods, can revolutionize the field of numerical linear algebra and change the world.

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