

Taming a white LED

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Invitation

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the public defence of
my PhD thesis title:

Taming a white LED

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TAMING A WHITE LED

EEN WITTE LED TEMMEN

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TAMING A WHITE LED

PROEFSCHRIFT

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To my father Leonid Borysovyh Meretskyi,
my first teacher of physics.

Contents

Table of Contents	vi
List of Figures	ix
Table of symbols and notations	xi
1 Introduction	1
1.1 Light	1
1.2 Diffusion of light	3
1.2.1 Single particle scattering	3
1.2.2 Diffusion of light	4
1.2.3 Diffusion of re-emitted light	5
1.3 Outline of the thesis	5
2 Radiative transfer equation (RTE): from diffusion theory to the P3 approximation	8
2.1 Introduction	8
2.2 PN approximation	8
2.3 Collimated incident beam as a source term	14
2.4 Source term for the re-emitted light	15
2.5 P1 and P3 approximations	16
2.6 Boundary conditions	20
2.7 Summary	22
3 How to distinguish elastically scattered light from Stokes-shifted light for solid-state lighting	23
3.1 Introduction	23
3.2 Theory	25
3.2.1 Scattered transmission with energy conversion	25
3.2.2 Scattered transmission in the absence of energy conversion	26
3.3 Experimental details	27
3.4 Results	30
3.4.1 Transmission measurements	30
3.4.2 Transport mean free path	33
3.4.3 Absorption mean free path	34
3.5 Summary and outlook	37
4 Accurate determination of transport parameters in strongly absorbing and scattering materials	39
4.1 Introduction	39

4.2	Model	41
4.3	Experimental details	44
4.4	Simulations	46
4.5	Results	47
4.5.1	Ballistic transmission	47
4.5.2	Diffuse light transmission and reflection	48
4.6	Discussion	53
4.7	Summary and outlook	54
5	Predicting the color of white LEDs	55
5.1	Introduction	55
5.2	Methods	57
5.3	Results and Discussion	58
5.4	White LED with backreflector	61
5.5	Summary	65
6	Universal limit on the applicability of diffusion theory for light propaga- tion	66
6.1	Introduction	66
6.2	Diffusion theory	66
6.3	Results	69
6.4	Error map of the diffusion theory	70
6.5	Summary	73
7	Reversible tuning of the color point of a white LED	75
7.1	Introduction	75
7.2	Wavefront shaping	75
7.3	Setup	79
7.4	Characterization of the setup	81
7.4.1	Optimization algorithm	81
7.4.2	Detection system	82
7.4.3	Measurement procedure	83
7.5	Results and discussion	85
7.6	Summary and outlook	87
8	Summary	89
	Nederlandse samenvatting	92
A	Color point	95
A.1	What is color?	95
A.2	Human eye	96
A.3	From spectrum to color	96
A.4	Color of objects	100
	Appendices	95
	References	102

List of Figures

1.1	Kyiv at night.	1
1.2	White LED.	2
1.3	Scattering and diffusion of light.	4
1.4	Scheme of the thesis.	6
2.1	Coordinate space used for radiative transfer equation.	9
3.2	Narrowband measurement setup.	29
3.3	Normalized reference spectra of the light sources used in the experiment. . .	29
3.4	Flux profile of the signal that we measure in the range where emission and absorption overlap for three different pump wavelengths.	30
3.5	Transport mean free path.	32
3.6	Determination of the absorption mean free path.	34
3.7	Absorption mean free path.	36
4.1	Transport parameters in the plane spanned by absorption and diffusion. . .	40
4.2	Scheme of light incident on and exiting from a slab.	42
4.3	Experimental setup for measuring transport parameters.	45
4.4	Ballistic transmission of the phosphor slab obtained in the experiment. . .	47
4.5	Measured scattering and absorption cross sections.	48
4.6	Transmission and scattered reflection spectra of silicone plates.	49
4.7	Transmission and reflection as a function of concentration for three chosen wavelengths.	50
4.8	Transport parameters of YAG:Ce ³⁺ phosphor particles extracted using ana- lytical model and Monte Carlo simulations.	52
5.1	Color point of a white LED.	56
5.2	Model for light propagation in a white LED slab.	56
5.3	Representative measured signal of a model white LED.	57
5.4	Transmission and reflection as a function of phosphor particle concentration.	59
5.5	Quantum yield of YAG:Ce ³⁺	61
5.6	White LED with back-reflector.	62
5.7	Total transmitted flux as a function of mirror reflectivity.	64
6.1	Validity range of diffusion theory for a slab shown as a 2D map.	68
6.2	Relative error in diffusion theory calculations shown as a 2D map.	71
6.3	Energy density (red curves) and flux (blue curves) in the unphysical range of transport parameters.	72

7.1	Tuning of the color point of a white LED.	76
7.2	Wavefront shaping technique.	77
7.3	Schematic of the diffused light cone inside a slab of scattering material. . .	78
7.4	A schematic of the setup for wavefront shaping on a phosphor sample. . . .	80
7.5	The wavefront enhancement registered on the CCD as a function of the number of optimization rounds N_{run}	81
7.6	Dark counts measurement for the experimental setup.	82
7.7	Calibration of the measurement setup.	83
7.8	Optimized spectra of the model white LED.	84
7.9	Enhancement as a function of target area for scattered and re-emitted light.	87
A.1	'The dress.'	95
A.2	Absorbance curve of the eye sensors.	96
A.3	The density of rods and cones in the eye.	97
A.4	Hue, saturation and luminance diagram.	98
A.5	Color matching experiment.	99
A.6	From color space $\bar{y}$ to color space Y	99
A.7	Spectrum of the white color D65.	100
A.8	Color vs illumination conditions.	101

Table of symbols and notations

Symbol	Units	Definition
σ_s	$\frac{1}{\text{mm wt}\% \text{ Hz}}$	scattering cross section
σ_a	$\frac{1}{\text{mm wt}\% \text{ Hz}}$	absorption cross section
σ_t	$\frac{1}{\text{mm wt}\% \text{ Hz}}$	total cross section
σ_{tr}	$\frac{1}{\text{mm wt}\% \text{ Hz}}$	transport cross section
μ	-	anisotropy factor
W	-	single particle albedo
ℓ_s	mm	scattering mean free path
ℓ_a	mm	absorption mean free path
ℓ_{tr}	mm	transport mean free path
b	-	optical thickness of the slab
λ	nm	wavelength
d	mm	thickness of the slab
ρ	$\#/\text{mm}^3$	number density of the scatterers
$T_{\text{tot}}^{\text{rel}}(\lambda)$	-	total relative transmission
T	-	total (elastic) transmission
R	-	total (elastic) reflection
T_b	-	ballistic transmission
T_r	-	re-emitted transmission
R_r	-	re-emitted reflection
U_d	$\frac{\text{W}}{\text{m}^2\text{sr Hz}}$	diffuse average intensity
F_0	$\frac{\text{W}}{\text{m}^2\text{Hz}}$	incident flux
η	-	enhancement

Chapter 1

Introduction

1.1 Light

Nearly all life on earth exists because of the extraterrestrial energy. Being the primary source of energy, the sun has defined and guided human lives for centuries. Even the moon would not shine in the sky if the sun disappeared or extinguished. Because the sun is so important for humans, a solar deity and sun worship can be found through most of the recorded history in many forms. A bonfire was perhaps the first artificial light source exploited by humans. Fire allowed humans to prepare food, and move around the planet at night, in brief, it greatly boosted human evolution [1].



Figure 1.1: **Kyiv at night.** Independence square ‘Maidan Nezalezhnosti’ in Ukraine at night. On the left side of the glass semi-sphere in the center are the Lach gates that were one of the entrances to the city. The first mention of the gates is traced to 1151.

From fire, people moved to more advanced light sources. The first commercially available light source that uses electricity was developed by Thomas Edison in 1878 [2]. This source

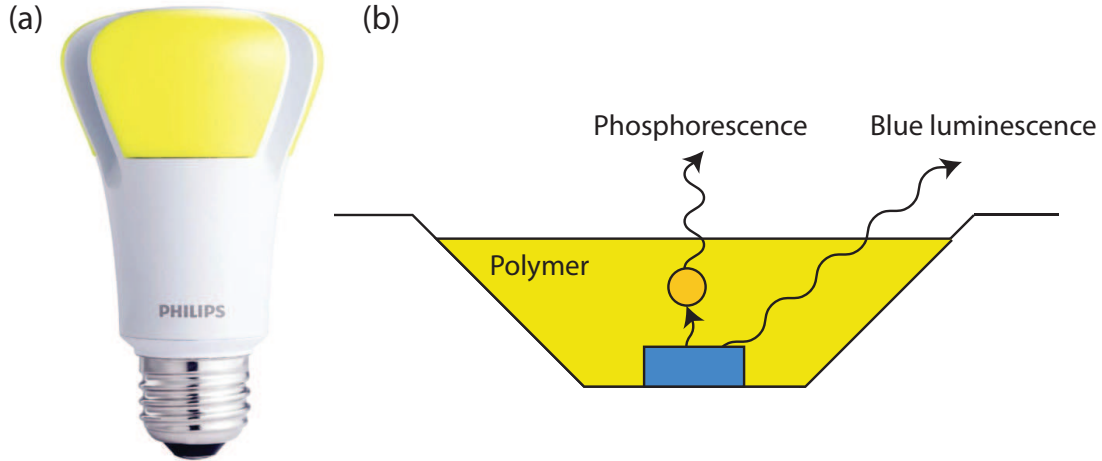


Figure 1.2: **White LED.** (a) L Prize winning LED lamp produced by Philips Lighting [14]. (b) Schematic of the basic construction of a white LED. A cup covered with aluminum is filled with phosphor particles that are mixed with the polymer. The phosphor particles absorb blue light emitted by the blue semiconductor and re-emit light in the red and green parts of the spectrum.

was an incandescent light bulb that can be found at homes even up to now. Currently, lighting is the second most energy consuming item in everyday life [3] (see Fig. 1.1¹). Since Edison's invention, the technology of white light sources has evolved tremendously. The Nobel prize-winning technology of the blue LED allowed to create energy-efficient white LEDs [4–8]. White LEDs are the most energy efficient light sources that can produce cold white light [9]. They are mechanically robust, thermally stable, possess excellent temporal stability, and have a long lifetime [9–12]. A typical white LED consists of a blue semiconductor LED [5,6,13] in combination with a phosphor layer [7,8] that consists of a dielectric matrix with a phosphor microparticles (see Fig. 1.2). A portion of the blue light is transmitted through the phosphor layer, and another portion is absorbed and re-emitted in the red and green part of the spectrum to yield the desired white light. The relative amount of a scattered and a re-emitted light defines the color point of a white LED that is explained in Appendix A.

In spite of the extensive use of solid-state lighting in everyday life and the apparent simplicity of the physical processes occurring in the phosphor layer, there is to date no analytical theory that predicts the spectrum of a white LED. The primary challenge arises from the lack of physical understanding of systems where multiple scattering and absorption

¹The L Prize is a prestigious prize established by the government. This award is aimed to stimulate lighting industry in developing high-quality and high-efficiency light sources.

of blue light coexist with the emission of light in a broadband wavelength range. Typically, numerical methods such as ray tracing and Monte Carlo techniques are used [15–18]. These methods do not have the predictive power of analytical theory. The numerical simulations require a set of heuristic parameters that are derived from the measurements on LEDs with a wide range of structural and optical parameters. The resulting heuristic parameters are often adjusted *a posteriori*, thereby further hampering the predictive power of these methods. Moreover, simulations are time-consuming and computationally demanding. All these aspects obstruct the efficient design of new white LEDs.

In this thesis, I present a radically new design principle for white LEDs. In chapter 1, I introduce the basic concepts required to describe light propagation in materials that are typically used for LEDs. In section 1.2, I describe the typical transport parameters used to define multiple light scattering, absorption, and re-emission in scattering materials. The outline of the thesis is given in section 1.3.

1.2 Diffusion of light

Traditionally, optics has been concerned with transparent components such as lenses, mirrors, and beam splitters [19], whereas so-called ‘dirty’ components that scatter light were avoided as much as possible. Over the years, however, the realization has arisen that spatial inhomogeneities that scatter light have advantageous properties and allow applications that are otherwise impossible, for instance, an optical diffuser or a high-numerical aperture objective [20–22]. Both the know-how of and the control over optics that strongly scatters light has significantly advanced [21–29]. However, the cutting edge is much less developed regarding optical systems that also strongly absorb light (or even re-emit light of a different color), even though essential application fields occur in this regime, for instance, solid-state lighting [8, 30, 31], biomedical optics [32–37], or powder technology [38–40]. In this section, I outline the transport parameters of a scattering material with absorption and re-emission that allow to describe the light propagation inside these materials.

1.2.1 Single particle scattering

When light illuminates a dielectric particle, part of the incident light is being scattered and absorbed by the particle (see Fig. 1.3 (a)). The amount of light that is scattered and absorbed is characterized by the scattering cross section σ_s and the absorption cross section σ_a . The scattering cross section σ_s is related to the total observed scattered power at all angles surrounding the particle. The absorption cross section σ_a is given by the total losses

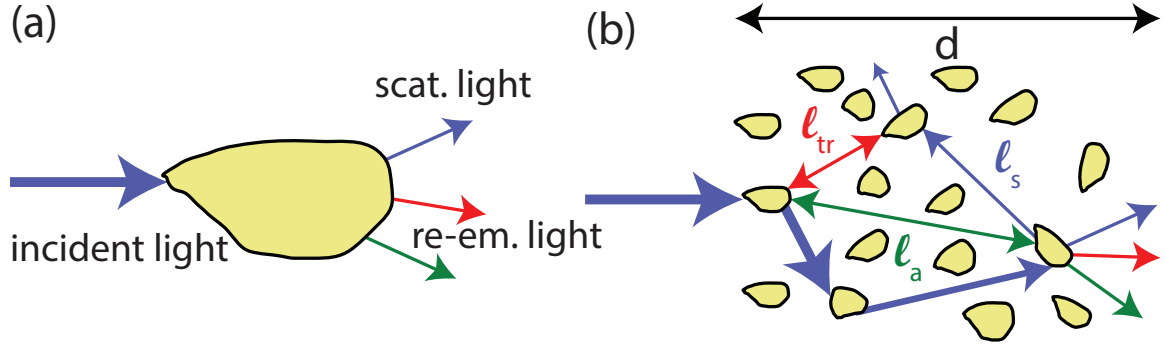


Figure 1.3: **Scattering and diffusion of light.** (a) A plane wave is incident upon a dielectric scatterer. The particle scatters, absorbs and re-emits incident light. (b) The multiple scattering medium with the characteristic length scales is shown. The size of the scattering medium is d , and the ℓ_s , ℓ_{tr} , ℓ_a are defined in the text.

of the power inside the dielectric particle. The sum of the scattering and the absorption cross sections is called the total cross section $\sigma_t = \sigma_s + \sigma_a$.

Depending on the size of the particle and the wavelength λ of the incident light the scattering process can be divided into three classes: Rayleigh scattering, Mie scattering, and geometrical-optics scattering. When the scattering particle is much smaller than the wavelength λ , Rayleigh scattering pertains. When the scattering particle size is comparable to the wavelength λ , Mie theory describes light scattering [41]. When the particle is much larger than the wavelength λ , geometrical optics applies.

1.2.2 Diffusion of light

Multiple scattering of light is typically described using the set of characteristic length scales ($d, \ell_s, \ell_a, \ell_{tr}$) that are shown in Fig. 1.3(b). The scattering mean free path ℓ_s is the mean distance between two consecutive scattering events. The absorption mean free path ℓ_a is the average diffuse propagation distance after which scattered light is absorbed by a fraction $(1/e)$. The transport mean free path ℓ_{tr} is the average distance after which light loses information about its original direction of propagation. These length scales are compared to the thickness of the scattering medium d to gauge the scattering regime.

When the interference effects inside the sample are negligible, the radiative transfer theory (RTE) describes the light propagation inside the scattering medium [42]. Radiative transfer theory is often simplified to diffusion theory when the following inequalities hold $\ell_{tr} \ll d \ll \ell_a$ (see Ch. 2) [25]. In most of the diffuser plates used for white LEDs, this is not the case. Instead, higher order approximations to the RTE should be used, for example, the P3 approximation - the third order approximation. The input set of the parameters for

the RTE and any order approximation is $(\rho, d, \sigma_s, \sigma_a, \mu)$, where ρ and μ are defined below.

Under the independent scattering assumption, the simple relation between the characteristic length scales $(d, \ell_s, \ell_a, \ell_{tr})$ and the single particle properties $(\sigma_s, \sigma_a, \mu)$ can be found, where μ is the anisotropy factor that characterizes the average cosine of the scattering angle θ of the particle. Hence, the input set of the parameters for the RTE, and any order approximation is $(\rho, d, \sigma_s, \sigma_a, \mu)$, where ρ is a number density of the scattering particles.

1.2.3 Diffusion of re-emitted light

In white LEDs, in addition to the scattering and absorption of light, the absorbed light is re-emitted. In the process of absorption, only a part of the incident light is being re-emitted. The rest of the energy is lost in the system in the form of heat. The efficiency of this process is characterized by the quantum yield $q(\lambda_1, \lambda_2)$, where λ_1 is the absorption wavelength and λ_2 is the emission wavelength. The quantum yield $q(\lambda_1, \lambda_2)$ is the ratio of the number of emitted photons to the number of absorbed photons.

The RTE is used to describe the propagation of the re-emitted light as in the previous case. However, I now need to know the input parameters $(\rho, d, \sigma_s, \sigma_a, \mu, q)$ at both the absorption and the re-emission wavelengths. Here, the absorbed light is taken to be the source of the re-emitted light with an efficiency q in the RTE for the re-emitted light.

1.3 Outline of the thesis

In this thesis, I experimentally and theoretically study the light propagating in phosphor diffusion layers that are typically used for white LEDs. Phosphor particles inside the phosphor layer scatter, absorb, and re-emit incident light. Absorption and scattering affect light in a comparable fashion. Hence, neither of the effects can be neglected. Therefore, widely used analytical tools fail to describe phosphor diffusion layers. I propose and experimentally verify the novel systematic approach that fully describes light diffusion and re-emission in phosphor layers. The proposed method can be used to analytically model the color point of a white LED, for the first time ever.

The connections between the thesis chapters are shown in Fig. 1.4. This thesis is organized in the following way: In Ch. 2, I derive the P1 and the P3 approximation to the radiative transfer equation (RTE) using a spherical-harmonics approach, to describe the diffusion of light in the phosphor layers. To describe the re-emitted light diffusion, I derive the P3 approximation for the re-emitted light, where the source of light is the absorbed diffused light. The derived expressions are used as an analytical foundation for describing

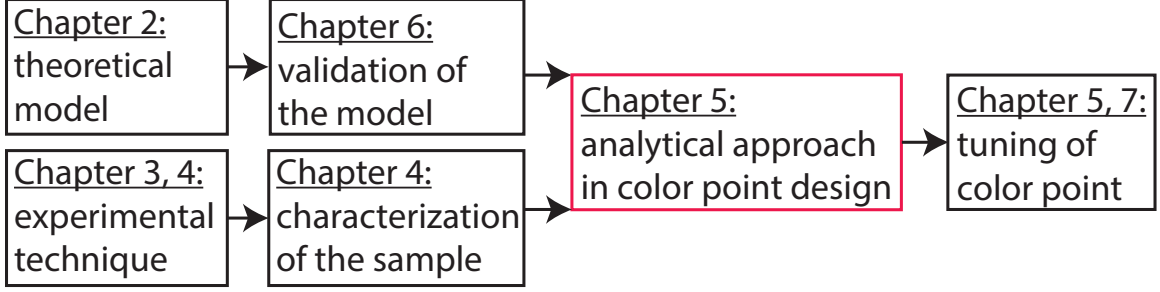


Figure 1.4: **Scheme of the thesis.** Arrows indicate the connections between the chapters of the thesis. The red box indicates the central question of the thesis.

light propagation and modeling color point of a white LED.

In Ch. 3, I develop the experimental technique that allows me to define transport parameters of the diffusers with strong absorption and re-emission in the whole visible spectral range. The standard experimental procedure fails in this case, because of the broadband light source that is used to measure the total transmission of the diffuser plates. When a broadband light source illuminates the diffuser with strong absorption and re-emission, the scattered and re-emitted light at the same wavelength cannot be spectrally separated. To achieve the separation of the different light components, I use a narrowband light source. A narrowband light source allows me to measure the transmission of light for the phosphor layer in the whole visible spectral range. I use diffusion theory (P1 approximation) to analyze experimental results. Diffusion theory connects measured transmission and the key transport parameters $(\ell_{\text{tr}}, \ell_{\text{a}})$ of the diffuser with absorption. From experimental data, I obtain one equation and two unknowns. To resolve the arising ambiguity, I assume that the transport mean free path ℓ_{tr} has a linear dependence on wavelength even in the strongly absorbing wavelength range of a phosphor. This assumption allows me to determine the set $(\ell_{\text{tr}}, \ell_{\text{a}})$ in the whole visible wavelength range.

In Ch. 4, I expand the experimental toolbox from Ch. 3. This expansion allows me to determine a wider set of the transport parameters $(\ell_{\text{tr}}, \ell_{\text{a}}, \mu)$ in the whole visible wavelength range for the scattering materials that are typically used for white LEDs. In this chapter, I use the P3 approximation to the radiative transfer equation (RTE) to analyze the experimental results. I find an excellent agreement between the transport parameters extracted using the P3 approximation and the Monte Carlo simulations. In contrast, the transport parameters extracted using the P1 approximation are off by $\sim 30\%$. The nature of this discrepancy is described in Ch. 6. The assumption regarding the linear dependence of the transport mean free path ℓ_{tr} made in Ch. 3 is confirmed.

In Ch. 5, I introduce a radically new design principle for white LEDs. My design

principle is based on laws of physics and predicts the central characteristics of a white LED, namely its color point. The proposed method allows exploring a large parameter space as it pertains to any phosphor and any substrate used in white LED production. My approach provides a deep understanding of white LEDs and results in fast and efficient design. This method realizes a reduction of both design and production costs of solid state lighting devices. It decreases the cost of ownership of solid state lighting devices and the consumption of expensive rare earth elements used by industry and increases the efficiency of solid state lighting devices. The proposed approach requires the knowledge of the average single-particle properties listed in Sec. 1.2, and extracted in Ch. 4.

In essence, my approach uses the P3 approximation to the RTE to describe the broadband re-emitted light. The input of the model is the set of transport parameters found in Ch. 4. The output of the model is the color of a white LED. This approach shows excellent agreement with presented experimental results and analytically maps a color point of a white LED to its spectrum. I identify the boundaries of physically achievable color points of white LED for the given design. I also experimentally extract the quantum yield q for the whole visible spectrum. It allows me to describe the re-emitted light propagation correctly.

In Ch. 6, I derive the universal limit on the validity of diffusion theory for light scattering media. This chapter is motivated by the inconsistencies of the scattering parameters found in Ch. 4, when the P1 or the P3 approximations are used. I derive analytical expressions that define the region of transport parameters where diffusion theory provides physical solutions. Outside this region diffusion theory gives apparent, yet erroneous, transport parameters.

In Ch. 7, I demonstrate the reversible color tuning of the color point of a white LED. I perform optical wavefront shaping to optimize the scattered or the re-emitted light in a large area ($\sim 1\text{mm}^2$) much larger than the wavelength squared. Remarkably, theoretical estimations fail to predict any enhancement in the transmission for such large areas. When optimizing either on the scattered or the re-emitted light, the color point of the emitted light shifts. When wavefront shaping is switched off, the color point of the white LED returns to the original position. My work demonstrates the possibility to extend wavefront shaping beyond nanophotonic systems to macroscopic systems. With the help of the developed technique, I can tune the color point of a white LED beyond the designed color point.

Chapter 2

Radiative transfer equation (RTE): from diffusion theory to the P3 approximation

2.1 Introduction

Transport theory deals, among others, with the transport of energy through a scattering medium. The radiative transfer equation (RTE) is the basic equation that is used in the transport theory of light [43]. Diffraction and interference effects are not included in the theory. It is assumed that there are no correlations between fields. Therefore, we consider the addition of powers rather than fields. Transport theory is equivalent to Boltzmann's equation used in the kinetic theory of gases in modern physics [43] or in the transport theory of neutrons. The radiative transfer equation is an integro-differential equation, and cannot be easily solved in the general case. In this chapter, we review the derivations of the most popular approximations such as the P1, also known as diffusion approximation, and the P3 approximation of the radiative transfer equation (RTE) in the slab geometry [43].

2.2 PN approximation

To derive the radiative transfer equation (RTE) we consider an elementary volume with unit height and length ds in space see Fig. 2.1 (a). The volume ds contains ρds particles, where ρ is the number density of the scattering particles. The specific intensity $I(\mathbf{r}, \hat{\mathbf{s}})$ is incident on this volume and is being scattered. The decrease of the specific intensity is written as

$$a = \frac{dI(\mathbf{r}, \hat{\mathbf{s}})}{ds} = -\rho\sigma_t I(\mathbf{r}, \hat{\mathbf{s}}). \quad (2.1)$$

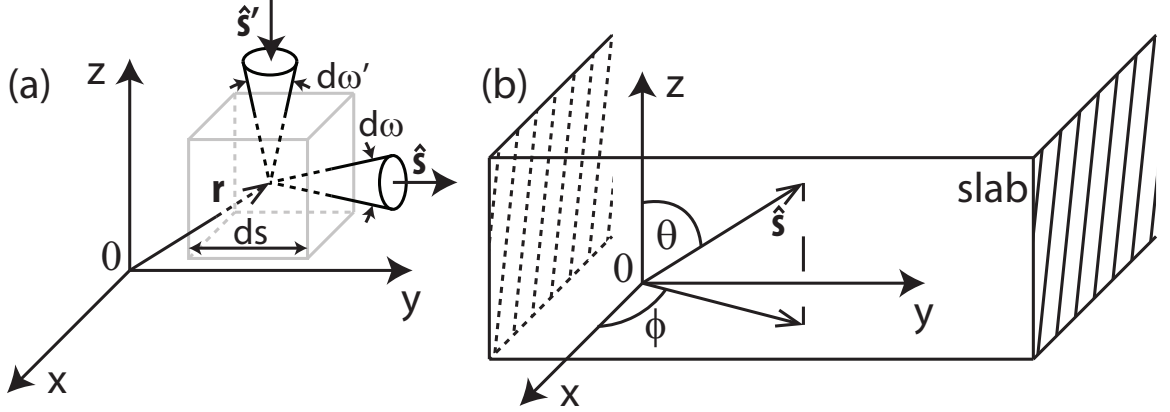


Figure 2.1: **Coordinate space used for radiative transfer equation.** (a) Coordinate system used in Eq. 2.3. (b) Coordinate system used in the Eq. 2.7 for the parallel slab geometry.

The specific intensity also increases when a portion of a specific intensity incident from an other direction $\hat{s}'$ is scattered into the direction $\hat{s}$

$$b = \frac{\rho\sigma_t}{4\pi} \int_{4\pi} p(\hat{s}, \hat{s}') I(\mathbf{r}, \hat{s}'), \quad (2.2)$$

where $p(\hat{s}, \hat{s}')$ is the phase function. The specific intensity can also increase due to the sources inside the medium. We sum all the contributions including the source terms to obtain the integral equation formulation of the RTE [43]:

$$\frac{dI_{\text{dif}}(\mathbf{r}, \hat{s})}{ds} = a + b + \text{sources} = -\rho\sigma_t I_{\text{dif}}(\mathbf{r}, \hat{s}) + \frac{\rho\sigma_t}{4\pi} \int_{4\pi} p(\hat{s}, \hat{s}') I_{\text{dif}}(\mathbf{r}, \hat{s}') d\omega' + \varepsilon_{\text{ri}}(\mathbf{r}, \hat{s}) + \varepsilon(\mathbf{r}, \hat{s}), \quad (2.3)$$

where $I_{\text{dif}}(\mathbf{r}, \hat{s})$ is the specific diffuse intensity (also known as the radiance or the brightness) that is the power per area per solid angle per bandwidth with units $[\frac{\text{W}}{\text{m}^2 \text{sr Hz}}]$, σ_t is the total cross section, $\varepsilon(\mathbf{r}, \hat{s})$ is the power generated per unit volume per unit solid angle in the direction $\hat{s}$ that accounts for all sources inside the medium, $\varepsilon_{\text{ri}}(\mathbf{r}, \hat{s})$ is the external source function, the power generated by the reduced intensity $I_{\text{ri}}(\mathbf{r}, \hat{s}')$

$$\varepsilon_{\text{ri}}(\mathbf{r}, \hat{s}) = \frac{\rho\sigma_t}{4\pi} \int_{4\pi} p(\hat{s}, \hat{s}') I_{\text{ri}}(\mathbf{r}, \hat{s}') d\omega', \quad (2.4)$$

and all vectors used in this equation are defined in Fig. 2.1(a). Here we expanded $I(\mathbf{r}, \hat{s})$ as

$$I(\mathbf{r}, \hat{s}) = I_{\text{ri}}(\mathbf{r}, \hat{s}) + I_{\text{dif}}(\mathbf{r}, \hat{s}), \quad (2.5)$$

where $I_{\text{dif}}(\mathbf{r}, \hat{\mathbf{s}})$ is the diffuse intensity and

$$\frac{dI_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}})}{ds} = -\rho\sigma_{\text{t}}I_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}) \quad (2.6)$$

is the reduced incident intensity.

In the parallel slab geometry the RTE becomes

$$\mu \frac{\partial I_{\text{dif}}(z, \mu)}{\partial z} = -\rho\sigma_{\text{t}}I_{\text{dif}}(z, \mu) + \frac{\rho\sigma_{\text{t}}}{4\pi} \int_{4\pi} p(\hat{\mathbf{s}}, \hat{\mathbf{s}}') I_{\text{dif}}(\mathbf{r}, \hat{\mathbf{s}}') d\omega' + \varepsilon_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}) + \varepsilon(\mathbf{r}, \hat{\mathbf{s}}), \quad (2.7)$$

where we use the equality $\frac{d}{ds} = \frac{\partial}{\partial z} \cdot \frac{dz}{ds} = \cos\theta \frac{\partial}{\partial z} = \mu \frac{\partial}{\partial z}$ [44], and the coordinate system is defined in Fig. 2.1(b).

To solve the RTE analytically, we expand every term in Eq. 2.7 in the basis of spherical harmonics [45]. This expansion allows us to separate variables $\hat{\mathbf{s}}$ and $\mathbf{r}$. Additionally, in many practical problems, rotational symmetries occur that can be exploited. For example, a scattering sample is illuminated with a collimated light beam with circular cross section, or the phase function $p(\hat{\mathbf{s}}, \hat{\mathbf{s}}')$ depends only on the difference between the incident and the scattered angles $\mu = \cos\theta = \hat{\mathbf{s}} \cdot \hat{\mathbf{s}}'^1$ [46]. Because of the symmetries, spherical harmonics are simplified to Legendre polynomials, since the dependence on the azimuth angle drops out (see Fig. 2.1(b)). As a first step, we will expand the phase function in the basis of Legendre polynomials and use addition theorem for spherical harmonics [47] to simplify the expression for the phase function $p(\hat{\mathbf{s}}, \hat{\mathbf{s}}')$

$$\begin{aligned} p(\hat{\mathbf{s}}, \hat{\mathbf{s}}') &= \sum_{k=0}^K \frac{2k+1}{4\pi} W_k P_k(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}') = \\ &= \sum_{k=0}^K \frac{2k+1}{4\pi} W_k \left[P_k(\mu) P_k(\mu') + \sum_{m=k}^K \frac{(k-m)!}{(k+m)!} P_k(\mu)^m P_k(\mu')^m \cos[m(\phi' - \phi)] \right], \end{aligned} \quad (2.8)$$

where W_k is the k -th moment of the phase function $p(\hat{\mathbf{s}}, \hat{\mathbf{s}}')$, $P_l(\hat{\mathbf{s}} \cdot \hat{\mathbf{s}}')$ is Legendre polynomial, $\mu = \cos\theta$ and $\mu' = \cos\theta'$ are the polar angles of the vectors $\hat{\mathbf{s}}$ and $\hat{\mathbf{s}}'$, ϕ and ϕ' are the azimuthal angles of the vectors $\hat{\mathbf{s}}$ and $\hat{\mathbf{s}}'$. The expansion is exact when $K \rightarrow \infty$. Further in the text, integration over the 4π angle produces zero value for the second term in Eq. 2.8.

¹This simplification arises due to the symmetries present in the system that eliminate the dependence on the angle ϕ .

We substitute the phase function $p(\hat{\mathbf{s}}, \hat{\mathbf{s}}')$ from Eq. 2.8 into the third term in Eq. 2.7

$$\begin{aligned} \frac{\rho\sigma_t}{4\pi} \int_{4\pi} p(\hat{\mathbf{s}}, \hat{\mathbf{s}}') I_{\text{dif}}(\mathbf{r}, \hat{\mathbf{s}}') d\omega' &= \frac{\rho\sigma_t}{4\pi} \int_{4\pi} I_{\text{dif}}(z, \mu') \sum_{k=0}^K (2k+1) W_k P_k(\mu) P_k(\mu') \sin\theta' d\theta' d\phi \\ &= 2\pi \frac{\rho\sigma_t}{4\pi} \int_{-1}^1 I_{\text{dif}}(z, \mu') \sum_{k=0}^K (2k+1) W_k P_k(\mu) P_k(\mu') d\mu'. \end{aligned} \quad (2.9)$$

In Eq. 2.9 the factor 2π comes from the integration over ϕ , the term proportional to $\cos[m(\phi' - \phi)]$ vanishes due to integration [32]. Eq. 2.7 becomes

$$\mu \frac{\partial I_{\text{dif}}(z, \mu)}{\partial z} = -\rho\sigma_t I_{\text{dif}}(z, \mu) + \rho\sigma_t \int_{-1}^1 p(\mu, \mu') I_{\text{dif}}(z, \mu') d\mu' + \varepsilon_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}) + \varepsilon(\mathbf{r}, \hat{\mathbf{s}}), \quad (2.10)$$

where

$$p(\mu, \mu') = \frac{1}{2} \sum_{k=0}^K (2k+1) W_k P_k(\mu) P_k(\mu'). \quad (2.11)$$

Next we expand the specific diffuse intensity in the basis of spherical harmonics [45]:

$$I_{\text{dif}}(\mathbf{r}, \hat{\mathbf{s}}) = \sum_{l=0}^L \sum_{m=-l}^l \psi_{lm}(\mathbf{r}) Y_{lm}(\hat{\mathbf{s}}), \quad (2.12)$$

where

$$\psi_{lm}(\mathbf{r}) = \int_{4\pi} I_{\text{dif}}(\mathbf{r}, \hat{\mathbf{s}}) Y_{lm}^*(\hat{\mathbf{s}}) d\hat{\mathbf{s}}, \quad (2.13)$$

and the spherical harmonics are defined as [48]

$$Y_{lm}(\hat{\mathbf{s}}) = \sqrt{\frac{2l+1}{4\pi} \frac{(l-m)!}{(l+m)!}} |\hat{\mathbf{s}}| P_l^m(\cos\theta) e^{im\phi}, \quad (2.14)$$

where $|\hat{\mathbf{s}}| = 1$, l is the degree of polynomial, m is the order of the polynomial, and the expansion is exact when $L \rightarrow \infty$. Because of the rotational symmetries present in the system, the spherical harmonics are simplified ($m = 0$). Diffuse intensity $I_{\text{dif}}(\mathbf{r}, \hat{\mathbf{s}})$ becomes:

$$I_{\text{dif}}(\mathbf{r}, \hat{\mathbf{s}}) = \sum_{l=0}^L \sum_{m=-l}^l \psi_{lm=0}(\mathbf{r}) Y_{lm=0}(\hat{\mathbf{s}}) = \sum_{l=0}^L \sqrt{\frac{2l+1}{4\pi}} \psi_l(\mathbf{r}) P_l(\cos\theta), \quad (2.15)$$

where in the second expression and further in the text we omit index m . To simplify the derivations we define

$$I_{\text{dif}}(\mathbf{r}, \hat{\mathbf{s}}) = \sum_{l=0}^L \frac{2l+1}{4\pi} \psi_l(\mathbf{r}) P_l(\cos\theta), \quad (2.16)$$

where

$$\psi_l(\mathbf{r}) = \int_{-1}^1 I_{\text{dif}}(\mathbf{r}, \hat{\mathbf{s}}) P_l(\mu) d\mu. \quad (2.17)$$

Until now we expanded the phase function $p(\hat{\mathbf{s}}, \hat{\mathbf{s}}')$ and the intensity $I_{\text{dif}}(\mathbf{r}, \hat{\mathbf{s}})$ in the Legendre polynomials basis. In the same way we expand the source term

$$\varepsilon_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}) = \sum_{l=0}^L \frac{2l+1}{4\pi} s_l(\mathbf{r}) P_l(\cos \theta), \quad (2.18)$$

where s_l is the l -th moment of the source function. We collect all the expanded terms and substitute Eq. 2.16 and Eq. 2.18 into Eq. 2.10 to obtain

$$\mu \frac{\partial I_{\text{dif}}(z, \mu)}{\partial z} + \sum_{l=0}^L \left[\rho \sigma_{\text{t}} \psi_l(z) P_l(\mu) - \rho \sigma_{\text{t}} \int_{-1}^1 p(\mu, \mu') \psi_l(z) P_l(\mu') d\mu' - s_l(\mathbf{z}) P_l(\mu) \right] \frac{2l+1}{4\pi} = 0. \quad (2.19)$$

This expression is the simplified RTE 2.3. It can be simplified even further when Eq. 2.19 is multiplied by $P_m(\mu)$ and integrated over 2π

$$\begin{aligned} & \int_{-1}^1 P_m(\mu) \mu \frac{\partial I_{\text{dif}}(z, \mu)}{\partial z} d\mu \\ & + \int_{-1}^1 d\mu \sum_{l=0}^L \frac{2l+1}{4\pi} \left[P_m(\mu) P_l(\mu) \rho \sigma_{\text{t}} \psi_l(z) - P_m(\mu) \rho \sigma_{\text{t}} \int_{-1}^1 p(\mu, \mu') \psi_l(z) P_l(\mu') d\mu' \right] \\ & = \int_{-1}^1 d\mu \sum_{l=0}^L \frac{2l+1}{4\pi} [P_m(\mu) P_l(\mu) s_l(\mathbf{z})]. \end{aligned} \quad (2.20)$$

In the next simplification step we use the following properties of the Legendre polynomials [46]. The term $P_m(\mu)$ in Eq. 2.9 can be expressed in the form

$$(2m+1)\mu P_m(\mu) = (m+1)P_{m+1}(\mu) + mP_{m-1}(\mu). \quad (2.21)$$

Another property of the Legendre polynomial that we use here is the orthogonality relation

$$\int_{-1}^1 P_m(\mu) P_l(\mu) d\mu = \frac{2}{2m+1} \delta_{ml}. \quad (2.22)$$

We start simplification with the first term on the left of Eq. 2.20. Substituting Eq. 2.21 results in

$$\begin{aligned} & \int_{-1}^1 P_m(\mu) \mu \frac{\partial I_{\text{dif}}(z, \mu)}{\partial z} d\mu = \int_{-1}^1 \left(\frac{m+1}{2m+1} P_{m+1}(\mu) + \frac{m}{2m+1} P_{m-1}(\mu) \right) \frac{\partial I_{\text{dif}}(z, \mu)}{\partial z} d\mu \\ & = \frac{m+1}{2m+1} \frac{\partial}{\partial z} \int_{-1}^1 P_{m+1}(\mu) I_{\text{dif}}(z, \mu) d\mu + \frac{m}{2m+1} \frac{\partial}{\partial z} \int_{-1}^1 P_{m-1}(\mu) I_{\text{dif}}(z, \mu) d\mu = \\ & = \frac{1}{2\pi} \left[\frac{m+1}{2m+1} \frac{d\psi_{m+1}(z)}{dz} + \frac{m}{2m+1} \frac{d\psi_{m-1}(z)}{dz} \right]. \end{aligned} \quad (2.23)$$

For the second term in Eq. 2.20 we use Eq. 2.22

$$\begin{aligned} \int_{-1}^1 d\mu \sum_{l=0}^L \frac{2l+1}{4\pi} P_m(\mu) P_l(\mu) \rho \sigma_t \psi_l(z) &= \sum_{l=0}^L \frac{2l+1}{4\pi} \rho \sigma_t \psi_l(z) \int_{-1}^1 P_m(\mu) P_l(\mu) d\mu \\ &= \sum_{l=0}^L \frac{2l+1}{4\pi} \rho \sigma_t \psi_l(z) \frac{2}{2m+1} \delta_{ml} = \frac{1}{2\pi} \rho \sigma_t \psi_m(z). \end{aligned} \quad (2.24)$$

For the third term in Eq. 2.20 we use the orthogonality relation Eq. 2.22

$$\begin{aligned} &\sum_{l=0}^L \frac{2l+1}{4\pi} \int_{-1}^1 d\mu P_m(\mu) \rho \sigma_t \int_{-1}^1 p(\mu, \mu') \psi_l(z) P_l(\mu') d\mu' \\ &= \frac{\rho \sigma_t}{2} \sum_{l=0}^L \frac{2l+1}{4\pi} \sum_{k=0}^K (2k+1) \int_{-1}^1 P_m(\mu) P_k(\mu) d\mu \int_{-1}^1 W_k P_k(\mu') \psi_l(z) P_l(\mu') d\mu' \\ &= \frac{2}{2m+1} \frac{\rho \sigma_t}{2} \sum_{l=0}^L \frac{2l+1}{4\pi} \sum_{k=0}^K (2k+1) \delta_{mk} \int_{-1}^1 W_k P_k(\mu') \psi_l(z) P_l(\mu') d\mu' \\ &= \frac{2}{2m+1} \frac{2m+1}{2} \rho \sigma_t \sum_{l=0}^L \frac{2l+1}{4\pi} \int_{-1}^1 W_m P_m(\mu') \psi_l(z) P_l(\mu') d\mu' \\ &= \frac{2\rho}{2m+1} \frac{2l+1}{4\pi} \delta_{ml} W_m \psi_l(z) = \frac{\rho \sigma_t}{2\pi} W_m \psi_m(z). \end{aligned} \quad (2.25)$$

The term on the right hand side in Eq. 2.20 is simplified with the orthogonality relation Eq. 2.22

$$\begin{aligned} \int_{-1}^1 d\mu \sum_{l=0}^L \frac{2l+1}{4\pi} [P_m(\mu) P_l(\mu) s_l(\mathbf{z})] &= \sum_{l=0}^L \frac{2l+1}{4\pi} s_l(\mathbf{z}) \int_{-1}^1 P_m(\mu) P_l(\mu) d\mu \\ &= \frac{1}{2\pi} s_m(\mathbf{z}). \end{aligned} \quad (2.26)$$

Adding all the simplified terms Eq. (2.23, 2.24, 2.25, 2.26) we simplify Eq. 2.20 to

$$\frac{m+1}{2m+1} \frac{d\psi_{m+1}(z)}{dz} + \frac{m}{2m+1} \frac{d\psi_{m-1}(z)}{dz} + \rho \sigma_t (1 - W_m) \psi_m(z) - s_m(\mathbf{z}) = 0, \quad (2.27)$$

where $m = 0, 1, 2, \dots, \infty$, and $\psi_{-1} = 0$. Next, we multiply Eq. 2.27 by $2m+1$, and arrive at the final expression

$$(m+1) \frac{d\psi_{m+1}(z)}{dz} + m \frac{d\psi_{m-1}(z)}{dz} + \rho \sigma_t (1 - W_m) (2m+1) \psi_m(z) - (2m+1) s_m(z) = 0. \quad (2.28)$$

Expression 2.28 is the infinite set of the coupled differential equations that describe light propagation in the scattering medium. To close this system of differential equations we make the P_M approximation, whereby we limit the expansion to order M . An odd positive

integer M is chosen with [35, 49]

$$\psi_m(z) = 0, \quad M < m < \infty. \quad (2.29)$$

The P_M approximation is usually expanded only to odd orders because even orders tend to be less accurate than the preceding odd order approximation [50, 51]. When M is chosen we are left with a system of $M + 1$ coupled first order differential equations, where $m = 0, 1, \dots, M$. When $M = 1$ the approximation is called $P1$ approximation. In this case the system of two coupled first order differential equations can be transformed to one second order differential equation. This second order differential equation is called diffusion equation (see Sec. 2.5).

2.3 Collimated incident beam as a source term

The source term in the radiative transfer equation is

$$\varepsilon_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}) = \frac{\rho\sigma_t}{4\pi} \int_{4\pi} p(\hat{\mathbf{s}}, \hat{\mathbf{s}}') I_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}') d\omega', \quad (2.30)$$

where the integration is performed over 4π solid angle. Let us consider a collimated incident beam as a source term. The incident intensity of the collimated beam in 1D slab geometry is equal to

$$I_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}') = F_0 \exp(-\rho\sigma_t z) \delta(\hat{\omega}' - \hat{\omega}_z), \quad (2.31)$$

where F_0 is the incident flux, $\hat{\omega}'$ and $\hat{\omega}_z$ are the solid angles, and $\hat{\omega}_z$ is aligned with the z axis in Fig. 2.1(b). We insert Eq. 2.31 into Eq. 2.30 for the source term, and we integrate over the angle ϕ to obtain the equation

$$\varepsilon_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}) = \frac{\rho\sigma_t}{4\pi} 2\pi \int_{-1}^1 I_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}') \sum_{l=0}^L (2l+1) W_l P_l(\mu) P_l(\mu') d\mu'. \quad (2.32)$$

We multiply this equation by $P_m(\mu)$, and integrate over 2π angle

$$\begin{aligned}
& \frac{\rho\sigma_t}{2} \int_{-1}^1 P_m(\mu) d\mu \int_{-1}^1 I_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}') \sum_{l=0}^L (2l+1) W_l P_l(\mu) P_l(\mu') d\mu' = \\
& \frac{\rho\sigma_t}{2} \frac{2}{2m+1} \int_{-1}^1 I_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}') \sum_{l=0}^L (2l+1) W_l P_l(\mu') \delta_{ml} d\mu' = \\
& \rho\sigma_t \int_{-1}^1 F_0 \exp(-\rho\sigma_t z) \delta(\hat{\omega}' - \hat{\omega}_z) W_m P_m(\mu') d\mu' = \\
& \rho\sigma_t F_0 \exp(-\rho\sigma_t z) W_m \int_{-1}^1 \delta(\hat{\omega}' - \hat{\omega}_z) P_m(\mu') d\mu' = \\
& \rho\sigma_t F_0 \exp(-\rho\sigma_t z) W_m P_m(\hat{\omega}_z) = \rho\sigma_t F_0 \exp(-\rho\sigma_t z) W_m P_m(1) = \\
& \rho\sigma_t F_0 \exp(-\rho\sigma_t z) W_m.
\end{aligned} \tag{2.33}$$

We substitute into Eq. 2.27 the following expression

$$s_m(z) = \rho\sigma_t F_0 \exp(-\rho\sigma_t z) W_m. \tag{2.34}$$

Coefficients W_m in Eq. 2.34 can be extracted by using the phase function $p(\mu, \mu')$ [43]

$$p(\mu, \mu') = \sum_{l=0}^L (2l+1) W_l P_l(\mu), \tag{2.35}$$

where

$$W_l = \int_{-1}^1 p(\mu) P_l(\mu) d\mu. \tag{2.36}$$

We define

$$p_1 = \frac{3}{4\pi} \int_{4\pi} p(\hat{\mathbf{s}}, \hat{\mathbf{s}}') \hat{\mathbf{s}} \cdot \hat{\mathbf{s}}' d\hat{\mathbf{s}}'. \tag{2.37}$$

Using Eq. 2.35 we get $p_1 = W_1$, $\mu = p_1/W_0 = W_1/W_0$, and finally $p_1 = W_1 = W_0\mu$, and $W_m = W_0\mu^m$, where $W_0 = \sigma_s/\sigma_t$ is the albedo of a single particle, and $\bar{\mu}$ is the mean cosine given by

$$\bar{\mu} = \left(\int_{4\pi} p(\hat{\mathbf{s}}, \hat{\mathbf{s}}') \mu d\hat{\mathbf{s}}' \right) / \left(\int_{4\pi} p(\hat{\mathbf{s}}, \hat{\mathbf{s}}') d\hat{\mathbf{s}}' \right). \tag{2.38}$$

2.4 Source term for the re-emitted light

In the previous section, the source of light was an reduced incident collimated beam with intensity $I_{\text{ri}}(\mathbf{r}, \hat{\mathbf{s}}')$. In white LEDs, light is not only scattered but also absorbed and re-emitted in the red and green parts of the spectra. The source function for the internally

re-emitted light is equal to [52]

$$\varepsilon(\mathbf{r}, \hat{\mathbf{s}}) = \varepsilon^{\text{re-em}}(\mathbf{r}, \hat{\mathbf{s}}) = \frac{\rho\sigma_t^{\text{ex}}}{4\pi} 2\pi \int_{-1}^1 I_a^{\text{ex}}(\mathbf{r}, \hat{\mathbf{s}}') \sum_{l=0}^L (2l+1) W_l P_l(\mu) P_l(\mu') d\mu', \quad (2.39)$$

where $\varepsilon^{\text{re-em}}(\mathbf{r}, \hat{\mathbf{s}})$ is the source function generated by the absorbed excitation light $I_a^{\text{ex}}(\mathbf{r}, \hat{\mathbf{s}}')$ and $\varepsilon_{\text{ri}}^{\text{re-em}}(\mathbf{r}, \hat{\mathbf{s}}) = 0$. We expand $I_a^{\text{ex}}(\mathbf{r}, \hat{\mathbf{s}}')$ in the same way as in Eq. 2.16, and after the repeating steps from Sec. 2.2 the source term becomes

$$s_m^{\text{re-em}}(z) = \rho\sigma_t^{\text{ex}} W_m^{\text{ex}} \psi_m^{\text{ex}}(z), \quad (2.40)$$

where $W_0^{\text{ex}} = \sigma_a^{\text{ex}}/\sigma_t^{\text{ex}}$, $W_m^{\text{ex}} = W_0 \mu_{\text{ex}}^m = 0$, the excitation light is being absorbed uniformly and hence $\mu_{\text{ex}}^m = 0$. When strong absorption is present in the system (see Ch. 6), we need to consider absorbed ballistic light and absorbed scattered light. The source term function then becomes a sum of two terms

$$s_m^{\text{re-em}}(z) = \rho\sigma_t^{\text{ex}} W_m^{\text{ex}} [\psi_m^{\text{ex}}(z)_{\text{dif}} + \psi_m^{\text{ex}}(z)_{\text{bal}}], \quad (2.41)$$

where $\psi_m^{\text{ex}}(z)_{\text{bal}} = F_0 \exp(-\rho\sigma_t^{\text{ex}} z)$, and $\psi_m^{\text{ex}}(z)_{\text{dif}}$ is a solution of the RTE for the scattered excitation light derived in Sec. 2.2.

2.5 P1 and P3 approximations

Let us consider the first order approximation to the RTE, namely the P1 approximation and compare it to the diffusion approximation $U_d(z)'' - \kappa_d^2 U_d(z) = -Q \exp(-\rho\sigma_t z)^2$ (where $U_d(z) = \frac{1}{4\pi} \psi_0$) derived in [43]. We take $m = 0, 1$ in Eq. 2.27

$$\frac{d\psi_1}{dz} + \rho\sigma_a \psi_0 = s_0, \quad (2.42)$$

$$\frac{d\psi_0}{dz} + 3\rho\sigma_{\text{tr}} \psi_1 = 3s_1, \quad (2.43)$$

where $\frac{d\psi_2}{dz} = 0$ (from Eq. 2.29), $s_1 = \rho\sigma_t F_0 \exp(-\rho\sigma_t z) W_1$, and $s_0 = \rho\sigma_t F_0 \exp(-\rho\sigma_t z) W_0$. This is the set of the first order coupled differential equations. From Eq. 2.43 we express ψ_1 through ψ_0 and s_1 as

$$\psi_1 = \frac{1}{3\rho\sigma_{\text{tr}}} (3s_1' - \psi_0'). \quad (2.44)$$

²In this section and further, symbol $'$ indicates a derivative over the spatial coordinate z .

We substitute this relation in Eq. 2.42 to obtain

$$\psi_0'' - \kappa_d^2 \psi_0 = -Q \exp(-\rho \sigma_t z), \quad (2.45)$$

where $\kappa_d^2 = 3(\rho \sigma_a)(\rho \sigma_{tr})$, $Q = (3\rho \sigma_s \rho \sigma_{tr} + 3\rho \sigma_s \rho \sigma_t \bar{\mu})F_0$. We find back the diffusion equation from [43], when we multiply Eq. 2.45 by $1/4\pi$

$$\frac{\partial^2}{\partial z^2} \left(\frac{1}{4\pi} \psi_0 \right) - \kappa_d^2 \left(\frac{1}{4\pi} \psi_0 \right) = -\frac{1}{4\pi} Q \exp(-\rho \sigma_t z) \Rightarrow (U_d(z) = \frac{1}{4\pi} \psi_0) \Rightarrow \quad (2.46)$$

$$\Rightarrow U_d(z)'' - \kappa_d^2 U_d(z) = -Q \exp(-\rho \sigma_t z), \quad (2.47)$$

where we redefined $Q = (3\rho \sigma_s \rho \sigma_{tr} + 3\rho \sigma_s \rho \sigma_t \bar{\mu})(F_0/4\pi)$ in Eq. 2.47.

When higher order approximations are considered, we solve the system of differential equations differently. We illustrate this derivation method by solving the P1 approximation. We start with the system of equations 2.42 and 2.43. There are two types of solutions for these equations: homogeneous and non-homogeneous. The non-homogeneous solution is

$$\psi_0^{nh} = G_0 \exp(-\rho \sigma_t z), \quad (2.48)$$

$$\psi_1^{nh} = G_1 \exp(\rho \sigma_t z), \quad (2.49)$$

where

$$G_0 = \frac{3(W_0 \gamma_1 \rho \sigma_t + W_1 \rho^2 \sigma_t^2)}{3\gamma_0 \gamma_1 - \rho^2 \sigma_t^2}, \quad (2.50)$$

$$G_1 = \frac{\rho \sigma_t (3W_1 \gamma_0 + W_0 \rho \sigma_t)}{3\gamma_0 \gamma_1 - \rho^2 \sigma_t^2}, \quad (2.51)$$

and

$$\gamma_0 = \rho \sigma_a, \quad (2.52)$$

$$\gamma_1 = \rho \sigma_{tr}. \quad (2.53)$$

The homogeneous solution is

$$\psi_0 = (H_{01} + H_{02})C_1 \exp(\mu_1 z), \quad (2.54)$$

$$\psi_1 = (H_{11} + H_{12})C_2 \exp(\mu_2 z), \quad (2.55)$$

where

$$H_{01} = 1, \quad (2.56)$$

$$H_{02} = 1, \quad (2.57)$$

$$H_{11} = -\frac{\gamma_0}{\mu_1}, \quad (2.58)$$

$$H_{12} = -\frac{\gamma_0}{\mu_2}, \quad (2.59)$$

and C_1, C_2 are integration constants. The full solution of the diffusion approximation to the radiative transfer equation becomes

$$\psi_m = \sum_{i=1}^2 H_{mi} C_i \exp(\mu_i z) + G_m \exp(-\rho \sigma_t z), \quad (2.60)$$

for $m = 0, 1$, where μ_i is the i -th root of the characteristic equation for the system of differential equations 2.42 and 2.43. The integration constants C_i are determined from the boundary conditions (see below).

We apply this solution method to find the solution to the next approximation of the RTE, namely the P3 approximation. To obtain the P3 approximation we take $m = 0, 1, 2, 3$, and $i = 1, 2, 3, 4$ in Eq. 2.60

$$\frac{d\psi_1}{dz} + \gamma_0 \psi_0 = \rho \sigma_t F_0 \exp(-\rho \sigma_t z) W_0, \quad (2.61)$$

$$2 \frac{d\psi_2}{dz} + \frac{d\psi_0}{dz} + 3\gamma_1 \psi_1 = 3\rho \sigma_t F_0 \exp(-\rho \sigma_t z) W_1, \quad (2.62)$$

$$3 \frac{d\psi_3}{dz} + 2 \frac{d\psi_1}{dz} + 5\gamma_2 \psi_2 = 5\rho \sigma_t F_0 \exp(-\rho \sigma_t z) W_2, \quad (2.63)$$

$$3 \frac{d\psi_2}{dz} + 7\gamma_3 \psi_3 = 7\rho \sigma_t F_0 \exp(-\rho \sigma_t z) W_3. \quad (2.64)$$

We take a homogeneous solution of the form

$$\psi_{mi}(z) = H_{mi} \exp(\mu_i z), \quad (2.65)$$

where $m = 0, 1, 2, 3$, and $i = 1, 2, 3, 4$, and insert it into Eq. (2.61, 2.62, 2.63, 2.64). The

equations (2.61, 2.62, 2.63, 2.64) in matrix form 2.64) become

$$\begin{pmatrix} \gamma_0 & \mu_i & 0 & 0 \\ \mu_i & 3\gamma_1 & 2\mu_i & 0 \\ 0 & 2\mu_i & 5\gamma_2 & 3\mu_i \\ 0 & 0 & 3\mu_i & 7\gamma_3 \end{pmatrix} \cdot \begin{pmatrix} H_{0i} \\ H_{1i} \\ H_{2i} \\ H_{3i} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \end{pmatrix}, \quad (2.66)$$

where $\gamma_m = \rho\sigma_t(1 - W_m)$, and $i = 1, 2, 3, 4$. The solution of the Eq. 2.75 is

$$H_{0i} = 1, \quad (2.67)$$

$$H_{1i} = -\frac{\gamma_0}{\mu_i}, \quad (2.68)$$

$$H_{2i} = \frac{3\gamma_1\gamma_0 - \mu_i^2}{2\mu_i^2}, \quad (2.69)$$

$$H_{3i} = \frac{-15\gamma_2\gamma_1\gamma_0 + 4\gamma_0\mu_i^2 + 5\gamma_2\mu_i^2}{6\mu_i^3}, \quad (2.70)$$

and the characteristic equation for μ_i becomes

$$9\mu_i^4 - \mu_i^2(35\gamma_2\gamma_3 + 27\gamma_1\gamma_0 + 28\gamma_3\gamma_0) + 105\gamma_0\gamma_1\gamma_2\gamma_3 = 0. \quad (2.71)$$

The solution of Eq. 2.71 contains four distinct roots, as it is the fourth order polynomial. The solution of Eq. 2.71 is

$$\mu_i = \pm \sqrt{(b \pm \sqrt{b^2 - 3780\gamma_0\gamma_1\gamma_2\gamma_3})/18}, \quad (2.72)$$

where $b = (35\gamma_2\gamma_3 + 27\gamma_1\gamma_0 + 28\gamma_3\gamma_0)$. We take an inhomogeneous solution of the form

$$\psi_m(z) = G_m \exp(-\rho\sigma_t z), \quad (2.73)$$

where $m = 0, 1, 2, 3$, and insert it into Eq. (2.61, 2.62, 2.63, 2.64). In matrix form Eq. (2.61,

2.62, 2.63, 2.64) become

$$\begin{pmatrix} \gamma_0 & -\rho\sigma_t & 0 & 0 \\ -\frac{\rho\sigma_t}{3} & \gamma_1 & \frac{2\rho\sigma_t}{3} & 0 \\ 0 & -\frac{2\rho\sigma_t}{5} & \gamma_2 & -\frac{3\rho\sigma_t}{5} \\ 0 & 0 & -\frac{3\rho\sigma_t}{7}\mu_i & \gamma_3 \end{pmatrix} \cdot \begin{pmatrix} G_0 \\ G_1 \\ G_2 \\ G_3 \end{pmatrix} = \begin{pmatrix} \frac{\rho\sigma_t}{2} F_0 W_0 \\ \frac{\rho\sigma_t}{2} F_0 W_1 \\ \frac{\rho\sigma_t}{2} F_0 W_2 \\ \frac{\rho\sigma_t}{2} F_0 W_3 \end{pmatrix}, \quad (2.74)$$

where $\gamma_m = \rho\sigma_t(1 - \frac{W_m}{2m+1})$. The solution of equation 2.74 becomes

$$\begin{pmatrix} G_0 \\ G_1 \\ G_2 \\ G_3 \end{pmatrix} = \begin{pmatrix} \frac{F_0\rho\sigma_t(105\gamma_1\gamma_2\gamma_3W_0 + \rho^2\sigma_t^2(-27\gamma_1W_0 - 28\gamma_3W_0 + 70\gamma_3W_2) + 105\gamma_2\gamma_3\rho\sigma_tW_1 + 3\rho^3\sigma_t^3(14W_3 - 9W_1))}{-2\rho^2\sigma_t^2(27\gamma_0\gamma_1 + 28\gamma_0\gamma_3 + 35\gamma_2\gamma_3) + 210\gamma_0\gamma_1\gamma_2\gamma_3 + 18\rho^4\sigma_t^4} \\ \frac{F_0\rho\sigma_t(-9\rho^3\sigma_t^3W_0 + 35\gamma_3\rho\sigma_t(\gamma_2W_0 + 2\gamma_0W_2) + 105\gamma_0\gamma_2\gamma_3W_1 + 3\gamma_0\rho^2\sigma_t^2(14W_3 - 9W_1))}{-2\rho^2\sigma_t^2(27\gamma_0\gamma_1 + 28\gamma_0\gamma_3 + 35\gamma_2\gamma_3) + 210\gamma_0\gamma_1\gamma_2\gamma_3 + 18\rho^4\sigma_t^4} \\ \frac{7F_0\rho\sigma_t(\gamma_3\rho^2\sigma_t^2(2W_0 - 5W_2) + 3\gamma_0\rho\sigma_t(2\gamma_3W_1 + 3\gamma_1W_3) + 15\gamma_0\gamma_1\gamma_3W_2 - 3\rho^3\sigma_t^3W_3)}{-2\rho^2\sigma_t^2(27\gamma_0\gamma_1 + 28\gamma_0\gamma_3 + 35\gamma_2\gamma_3) + 210\gamma_0\gamma_1\gamma_2\gamma_3 + 18\rho^4\sigma_t^4} \\ \frac{F_0\rho\sigma_t(3\rho^3\sigma_t^3(2W_0 - 5W_2) + \rho^2\sigma_t^2(18\gamma_0W_1 - 7W_3(4\gamma_0 + 5\gamma_2)) + 45\gamma_0\gamma_1\rho\sigma_tW_2 + 105\gamma_0\gamma_1\gamma_2W_3)}{-2\rho^2\sigma_t^2(27\gamma_0\gamma_1 + 28\gamma_0\gamma_3 + 35\gamma_2\gamma_3) + 210\gamma_0\gamma_1\gamma_2\gamma_3 + 18\rho^4\sigma_t^4} \end{pmatrix}, \quad (2.75)$$

The final solution of Eq. (2.61, 2.62, 2.63, 2.64) is obtained by combining the homogeneous and the inhomogeneous solutions.

2.6 Boundary conditions

In the P1 approximation or diffusion approximation, the net flux of light is conserved across the boundaries of the slab. This conservation law results in two boundary conditions [33]

$$\int_0^1 I_{\text{dif}}(0, \mu) P_l(\mu) d\mu = \int_0^1 R_2(\mu) I_{\text{dif}}(0, -\mu) P_l(\mu) d\mu, \quad (2.76)$$

$$\int_{-1}^0 I_{\text{dif}}(d, -\mu) R_2(\mu) P_l(\mu) d\mu = \int_{-1}^0 I_{\text{dif}}(d, \mu) P_l(\mu) d\mu, \quad (2.77)$$

where $l = 1$, and $R_2(\mu)$ is the specular Fresnel reflection function for unpolarized light. We substitute the expression for intensity Eq. 2.16 in Eq. 2.77 that results in

$$\sum_{m=0}^M \frac{2m+1}{4\pi} \psi_m(0) \int_0^1 P_m(\mu) P_l(\mu) d\mu = \sum_{m=0}^M \frac{2m+1}{4\pi} \psi_m(0) \int_0^1 R_2(\mu) P_m(-\mu) P_l(\mu) d\mu, \quad (2.78)$$

$$\sum_{m=0}^M \frac{2m+1}{4\pi} \psi_m(d) \int_{-1}^0 R_2(\mu) P_m(-\mu) P_l(\mu) d\mu = \sum_{m=0}^M \frac{2m+1}{4\pi} \psi_m(d) \int_{-1}^0 P_m(\mu) P_l(\mu) d\mu. \quad (2.79)$$

For the P1 approximation $M = 1$, and the boundary conditions 2.78 and 2.79 become

$$\psi_0(0) \int_0^1 P_0(\mu) P_1(\mu) (1 - R_2(\mu)) d\mu + 3\psi_1(0) \int_0^1 P_1(\mu) P_1(\mu) (1 + R_2(\mu)) d\mu = 0, \quad (2.80)$$

$$\psi_0(d) \int_{-1}^0 P_0(\mu) P_1(\mu) (1 - R_2(\mu)) d\mu + 3\psi_1(d) \int_{-1}^0 P_1(\mu) P_1(\mu) (1 + R_2(\mu)) d\mu = 0. \quad (2.81)$$

For the P3 approximation, we need two more equations. These additional equations are obtained by taking $l = 3$ and $M = 3$ in Eq. 2.78 and 2.79.

$$\sum_{m=0}^3 \frac{2m+1}{4\pi} \psi_m(0) \int_0^1 P_m(\mu) P_3 d\mu = \sum_{m=0}^3 \frac{2m+1}{4\pi} \psi_m(0) \int_0^1 R_2(\mu) P_m(-\mu) P_3 d\mu, \quad (2.82)$$

$$\sum_{m=0}^3 \frac{2m+1}{4\pi} \psi_m(d) \int_{-1}^0 R_2(\mu) P_m(-\mu) P_3 d\mu = \sum_{m=0}^3 \frac{2m+1}{4\pi} \psi_m(d) \int_{-1}^0 P_m(\mu) P_3 d\mu, \quad (2.83)$$

where $P_0 = 1$, $P_1 = \mu$, $P_2 = (3\mu^2 - 1)/2$, $P_3 = (5\mu^3 - 3\mu)/2$. The boundary conditions for the P3 approximation become

$$J_0 \psi_0(0) + 3J_1 \psi_1(0) + 5J_2 \psi_2(0) + 7J_3 \psi_3(0) = 0, \quad (2.84)$$

$$D_0 \psi_0(d) + 3D_1 \psi_1(d) + 5D_2 \psi_2(d) + 7D_3 \psi_3(d) = 0, \quad (2.85)$$

$$E_0 \psi_0(0) + 3E_1 \psi_1(0) + 5E_2 \psi_2(0) + 7E_3 \psi_3(0) = 0, \quad (2.86)$$

$$F_0 \psi_0(d) + 3F_1 \psi_1(d) + 5F_2 \psi_2(d) + 7F_3 \psi_3(d) = 0, \quad (2.87)$$

where

$$J_0 = -D_0 = \int_0^1 \mu (1 - R(\mu)) d\mu, \quad (2.88)$$

$$J_1 = D_1 = \int_0^1 \mu^2 (1 + R(\mu)) d\mu, \quad (2.89)$$

$$J_2 = -D_2 = \int_0^1 \mu \frac{3\mu^2 - 1}{2} (1 - R(\mu)) d\mu, \quad (2.90)$$

$$J_3 = D_3 = \int_0^1 \mu \frac{5\mu^3 - 3\mu}{2} (1 + R(\mu)) d\mu, \quad (2.91)$$

and

$$E_0 = -F_0 = \int_0^1 \frac{5\mu^3 - 3\mu}{2} (1 - R(\mu)) d\mu, \quad (2.92)$$

$$E_1 = F_1 = \int_0^1 \mu \frac{5\mu^3 - 3\mu}{2} (1 + R(\mu)) d\mu, \quad (2.93)$$

$$E_2 = -F_2 = \int_0^1 \frac{5\mu^3 - 3\mu}{2} \frac{3\mu^2 - 1}{2} (1 - R(\mu)) d\mu, \quad (2.94)$$

$$E_3 = F_3 = \int_0^1 \frac{5\mu^3 - 3\mu}{2} \frac{5\mu^3 - 3\mu}{2} (1 + R(\mu)) d\mu, \quad (2.95)$$

where $R(\mu)$ is Fresnel reflection coefficient for unpolarised light.

2.7 Summary

In this chapter, I review two major steps in deriving the P1 and the P3 approximation in the slab geometry. The first step is to expand all the terms of the RTE in the spherical harmonics and decouple $\mathbf{r}$ and $\hat{\mathbf{s}}$. Next step is to reduce the dimensionality of the equation. I explicitly show how P1 approximation can be reduced to the well known form of the diffusion equation. The presented derivations can be found throughout the scattered literature [32,33, 35,43–51]. In this chapter, these derivations are collected and presented with the consistent notations. The derived expressions account for the boundary conditions. The expression for the re-emitted light in the P3 approximation is derived considering the solution of the P3 approximation to the scattered light as a source function. The derived expressions are used throughout this thesis.

Chapter 3

How to distinguish elastically scattered light from Stokes-shifted light for solid-state lighting

3.1 Introduction

An important class of white light sources is the white LED that possess numerous advantages over conventional sources, such as incandescent lamps or discharge lamps. White LEDs are the most energy-efficient sources. They are mechanically robust and thermally stable, they possess excellent temporal stability, and a long lifetime [9, 10]. A typical white LED consists of a blue semiconductor LED in combination with a phosphor layer that consists of a dielectric matrix with the phosphor microparticles [8]. To describe the light propagation in white LEDs, we must define the transport parameters of the phosphor layer. Previously a broadband light source was applied to obtain optical properties of the diffuser plates [53], and phosphor plates [54]. This approach fails when the re-emitted light overlaps spectrally with the elastically scattered light. In Fig. 3.1(c), we show the overlap region for the plates studied here in a $x - y$ chromaticity diagram. This spectral range corresponds to the green part of the white LED spectrum where the human eye is most sensitive [55]. Here, we present a novel measurement technique that allows us to separate the elastically scattered and Stokes-shifted light in the overlap range for phosphor diffuser plates that are used in commercial white LEDs. As a result, we now close the "the green gap" in Fig. 3.1(c) and extract the relevant transport and absorption parameters for solid-state lighting in the whole visible spectral range.

We use analytical theory originating from nanophotonics, wherein propagation of light is described from first principles [23, 26, 56–58]. Such an *ab initio* theory supplies fundamental physical insights on the light propagation inside solid-state lighting device [53, 54].

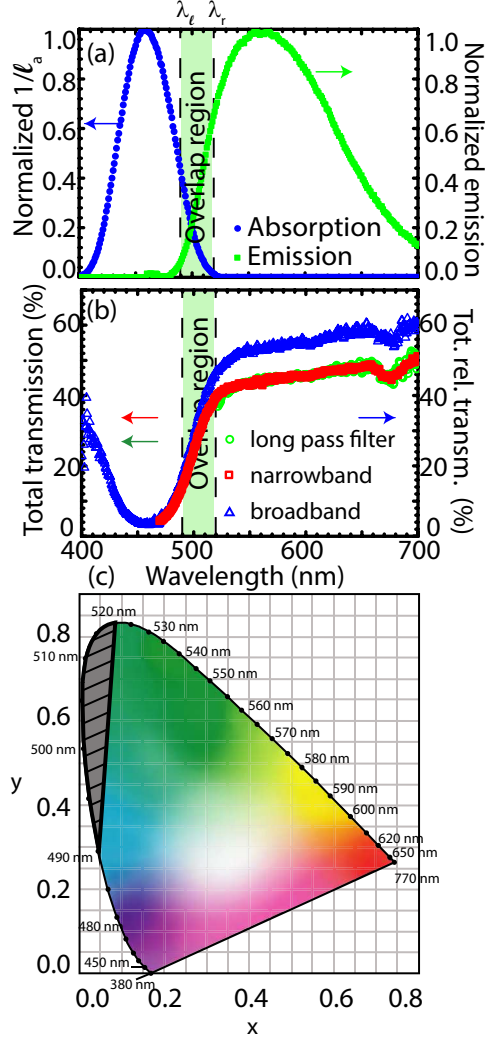


Figure 3.1: **(a)** Normalized absorption spectrum (blue circles) and emission spectrum (green squares) of the YAG:Ce³⁺ phosphor that is used in our study are shown. The spectral range where emission and absorption spectra overlap is indicated with a green bar between λ_ℓ and λ_r . **(b)** Transmission spectrum obtained by using the narrowband (red squares) and the broadband (green circles) light sources for the polymer plate with 4 wt % phosphor particles. Arrows point to the relevant ordinate for the data. **(c)** CIE 1931 (x,y) chromaticity diagram [8]. Monochromatic colors are located at the perimeter of the diagram. The white color is located in the middle of the diagram. The hatched gray area represents the region where emission and absorption of YAG:Ce³⁺ overlap. The overlap range was previously inaccessible, and it is made accessible in this work.

Extracting the optical parameters from theory is less time consuming than performing many simulations. More importantly: the resulting parameters are robust, and predictions can be made beyond the parameter range that was initially studied. For instance, knowledge of the absorption spectra provides us with the design guidelines for the solid-state light units. The design parameters such as the thickness of the diffuser plates and the phosphor

concentration can be directly extracted from the absorption spectra depending on the blue pump wavelength of a white LED.

3.2 Theory

3.2.1 Scattered transmission with energy conversion

Multiple light scattering is usually studied by measuring the scattered transmission through a slab of a complex, multiple scattering medium [59,60]. Scattered transmission, or diffuse transmission, is the transmission of an incident collimated beam with flux $F_0(\lambda_i)$ at the incident wavelength λ_i that is multiply scattered and integrated over all outgoing angles of the light exiting from a medium in the forward direction. The scattered transmission carries information on the transport mean free path ℓ_{tr} and on the absorption mean free path ℓ_a , which are the crucial parameters that describe multiple light scattering [26,43,61–64]. The transport mean free path ℓ_{tr} is the distance it takes for the direction of light to become randomized while performing a random walk in a scattering medium. The absorption mean free path ℓ_a is the distance it takes for light to be absorbed by a fraction $(1/e)$ while light performs a random walk in a scattering medium.

Phosphor particles do not only scatter light, but also convert blue light into other colors by absorbing blue and re-emitting other colors of light. Therefore, from here on we will refer to the measured scattered transmission in presence of energy conversion as the *total relative transmission* $T_{tot}^{rel}(\lambda)$

$$T_{tot}^{rel}(\lambda) = \frac{F_{tot}(\lambda_i)}{F_0(\lambda_i)}, \quad (3.1)$$

where $F_{tot}(\lambda_i)$ is the transmitted flux that is collected at the forward side of the diffusion plate. In the emission range of a phosphor ($\lambda \geq \lambda_\ell$) the collected flux $F_{tot}(\lambda_i)$ can be written as a sum of the elastic transmitted flux $F_T(\lambda_i)$ and the transmitted Stokes-shifted flux $F_{em}(\lambda_i)$. Thus, the total relative transmission $T_{tot}^{rel}(\lambda)$ can be separated into two parts [54]

$$T_{tot}^{rel}(\lambda_i) = T(\lambda_i) + T_{em}(\lambda_i) = \frac{F(\lambda_i) + F_{em}(\lambda_i)}{F_0(\lambda_i)}, \quad (3.2)$$

where the first term $T(\lambda_i)$ is the total elastic transmission, and the second term $T_{em}(\lambda_i)$ is the re-emitted light that experienced the energy conversion in the diffuse absorptive medium. In Ref. [54] these two terms could not be distinguished in the overlap range $\lambda_\ell < \lambda < \lambda_r$.

The central question in this chapter is how to distinguish the scattered transmission $T(\lambda_i)$ from the total relative transmission T_{tot}^{rel} as this allows one to obtain both the transport mean free path ℓ_{tr} and the absorption mean free path ℓ_a . To access the scattered

transmission, we employ a tunable narrowband light source and spectrally resolve the narrowband transmitted light. Since the light that is converted by the phosphor exhibits a Stokes-shift $F_{\text{em}}(\lambda_i)$, this part $T_{\text{em}}(\lambda_i)$ of the total relative transmission is filtered. Hence, we obtain the desired total elastic transmission $T(\lambda_i)$ that we interpret with diffusion theory.

3.2.2 Scattered transmission in the absence of energy conversion

According to diffusion theory for light, the scattered transmission $T(\lambda_i)$ through a slab, even in the presence of absorption, is a function of the slab thickness d , the wavelength λ , the transport mean free path ℓ_{tr} , and the absorption ℓ_{a} mean free path, and can be expressed as [43]¹:

$$T(d, \lambda_i, \ell_{\text{tr}}, \ell_{\text{a}}) = Q^{-1} [\sinh(\mu_{\text{a}} z_p) + \mu_{\text{a}} z_{\text{e}} \cosh(\mu_{\text{a}} z_p)], \quad (3.3)$$

with

$$Q(d, \lambda_i, \ell_{\text{tr}}, \ell_{\text{a}}) \equiv (1 + \mu_{\text{a}}^2 z_{\text{e}}^2) \sinh(\mu_{\text{a}} d) + 2\mu_{\text{a}} z_{\text{e}} \cosh(\mu_{\text{a}} d), \quad (3.4)$$

where the extrapolation lengths are equal to

$$z_{e1}(\ell_{\text{tr}}) = z_{e2}(\ell_{\text{tr}}) = z_{\text{e}}(\ell_{\text{tr}}) = \frac{2}{3} \ell_{\text{tr}} \frac{1 + \overline{R}_{1,2}}{1 - \overline{R}_{1,2}}. \quad (3.5)$$

Here z_p is the diffuse penetration depth of light, $\mu_{\text{a}} \equiv 1/\ell_{\text{a}}$ the inverse absorption mean free path, $\overline{R}_{1,2}$ is the angular and polarization averaged reflectivity of the respective boundaries [65]. For a normal incident collimated beam the penetration depth becomes $z_p = \ell_{\text{tr}}$ [66], and $\overline{R}_{1,2} = 0.57$ for polymer plates with an average refractive index $n = 1.5$ from Ref. [67]. For samples with no absorption ($\mu_{\text{a}} = 0$), Eq. (3.3) simplifies to the optical Ohm's law [23]

$$\lim_{\ell_{\text{a}} \rightarrow \infty} T(d, \lambda_i, \ell_{\text{tr}}, \ell_{\text{a}}) = T(d, \lambda_i, \ell_{\text{tr}}) = \frac{\ell_{\text{tr}} + z_{\text{e}}}{d + 2z_{\text{e}}}. \quad (3.6)$$

In the range of zero phosphor absorption, the scattered transmission is a function of the sample thickness d , the incoming wavelength λ_i , and the transport mean free path ℓ_{tr} , $T = T(d, \lambda_i, \ell_{\text{tr}})$. Therefore, we can extract ℓ_{tr} using Eq. (3.6) from measurements of the scattered transmission $T(\lambda_i)$ in the range of no absorption ($\lambda_i \geq \lambda_r$). In the range of

¹In Ref. [54] an approximation was used that is only valid for low absorption. Here we use the exact expression hence our results are also valid for strong absorption (high μ_{a}). As a result, our present results differ from Ref. [54] in the spectral range where strong absorption appears, notably near the peak of the phosphor absorption.

strong light absorption ($\lambda_i \leq \lambda_r$) the scattered transmission also depends on the absorption mean free path ℓ_a : $T = T(d, \lambda_i, \ell_{tr}, \ell_a)$. Therefore, the transport mean free path ℓ_{tr} has to be derived separately. We extrapolate the ℓ_{tr} values, extracted in the zero absorption wavelength range ($\lambda_i \geq \lambda_r$), into the strong absorption wavelength range. It is possible because ℓ_{tr} is a monotonously increasing function of λ for size-polydisperse scatterers [53, 58, 68]. By measuring the scattered transmission in the range of strong absorption, we thus obtain ℓ_a using extrapolated values of ℓ_{tr} , by using Eq. (3.3). The central question in this chapter is how to distinguish the scattered transmission $T(\lambda_i)$ from the total relative transmission T_{tot}^{rel} as this allows one to obtain both the transport mean free path ℓ_{tr} and the absorption mean free path ℓ_a . To access the scattered transmission, we employ a tunable narrowband light source and spectrally resolve the narrowband transmitted light. Since the light that is converted by the phosphor exhibits a Stokes-shift $F_{em}(\lambda_i)$, this part $T_{em}(\lambda_i)$ of the total relative transmission is filtered. Hence, we obtain the desired total elastic transmission $T(\lambda_i)$ that we interpret with diffusion theory.

3.3 Experimental details

We have studied the light transport through polymer plates that are used in Fortimo solid-state lighting units [69]. The polymer plates consist of a polycarbonate matrix (Lexan 143R) with YAG:Ce³⁺ ceramic phosphor particles that are widely used in white LEDs. The phosphor particles have a broad diameter distribution centered around 10 μm [54], and a Ce³⁺ concentration in the YAG:Ce³⁺ of 3.3 wt %.

The emission and absorption spectra of the YAG:Ce³⁺ are shown in Fig. 3.1(a) for the powder that was used in the polymer diffuser plates. The absorption and emission bands have peaked at 458 nm and 557 nm, respectively, and overlap in the spectral range between $\lambda_\ell = 490$ nm and $\lambda_r = 520$ nm. As a result, we distinguish three spectral ranges in the visible spectrum where different physical processes are taking place: (1) in the spectral range up to $\lambda_\ell = 490$ nm, light is partly elastically scattered and partially absorbed. This range has already been studied in Ref. [54]. (2) In the spectral range between $\lambda_\ell = 490$ nm and $\lambda_r = 520$ nm, incident light is elastically scattered and absorbed inside the sample. The absorbed light is re-emitted by the phosphor, and subsequently elastically scattered. This is the overlap range that is central to the present work. (3) In the spectral range beyond $\lambda_r = 520$ nm, the emitted light is the sum of the incident light that is elastically scattered and re-emitted light by the phosphor that is also elastically scattered. This range has also already been studied in Ref. [54].

In this chapter, we present the transmission measurements on five such polymer plates with a phosphor concentration ranging from $\phi = 2.0$ to 4.0 wt%. The corresponding volume fractions range from $\phi_{\text{vol}} = 0.5$ to 1 vol%, which is in the limit of low scatterer concentration because the transport mean free path is of the order of sample thickness for the given volume fractions. The plates were prepared using injection molding where a powder of YAG:Ce³⁺ particles is mixed with the polymer powder, and the mixture is melted and pressed into a press-form. The polymer plates, shown in Fig. 3.2(a), are circular with a diameter of 60 mm and a thickness of 2 mm.

Figure 3.2(b) shows a drawing of the setup for measuring the spectrally resolved scattered transmission $T(\lambda, d, \rho)$, where ρ is the number density of the scattering particles, with a narrowband incident light beam. We illuminate the sample with two different light sources: a tunable narrowband light source and a broadband light source. The narrowband light source consists of a Fianium supercontinuum white-light source (WL-SC-UV-3), which is spectrally filtered to a bandwidth of less than $\Delta\lambda = 2.4$ nm by a prism monochromator (Carl-Leiss Berlin-Steglitz). The wavelength of the narrowband source is tunable in the wavelength range between 400 and 700 nm, as shown in Fig 3.3. The infrared light above 700 nm of the supercontinuum laser source is filtered by a neutral density filter (NENIR30A) and a dichroic mirror (DMSP805). The spectrum of the supercontinuum source after filtering the infrared light is shown in Fig. 3.3. The spectrum varies as a function of wavelength. These variations are reproducible so that we can normalize measured spectrum.

The incident beam illuminates the phosphor plate at normal incidence, and the plate is placed at the entrance port of an integrating sphere. We verified that the entrance port of the integrating sphere is sufficiently large to collect all flux that emanated from the strongest scattering sample. The flux of the outgoing light entering the integrating sphere is analyzed with a fiber-to-chip spectrometer (AvaSpec-USB2-ULS2048L) with a spectral resolution of $\Delta\lambda = 2.4$ nm.

An example of the measured spectra for three incident wavelengths $\lambda_i = 527, 550$ and 634 nm are shown in Fig. 3.3 with the red squares and red dotted lines. The peak flux varies significantly across the spectral range as a result of the spectral variation of the supercontinuum source. To maintain a fixed reference flux, we automatically adjust the integration time at every wavelength during measurements.

The alternative broadband light source consist of a white LED (Luxeon LXHL-MW1D) (see Fig. 3.2) with an emission spectrum covering the range from 400 nm to 700 nm as shown in Fig. 3.3. This source is not filtered.

For all phosphor plates, we measured the transmission spectra with both the narrow-

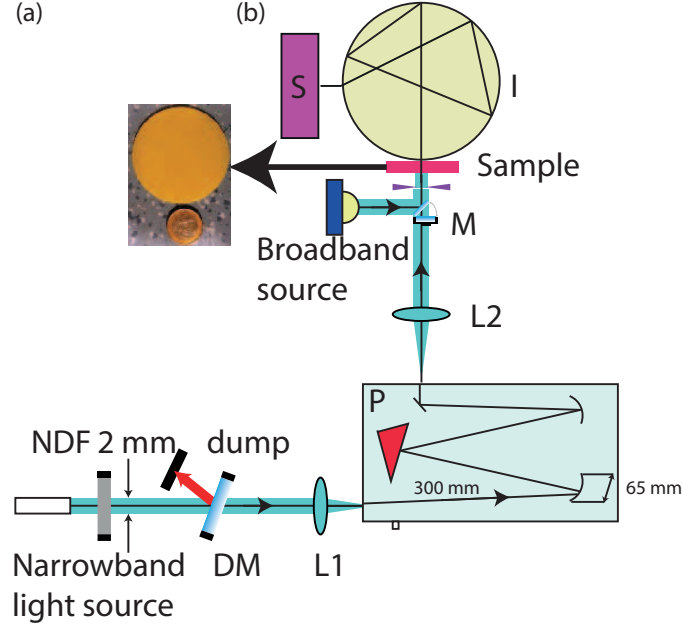


Figure 3.2: **Narrowband measurement setup.** (a) A polymer slab with a 4 wt% YAG:Ce³⁺ compared to a 1 Euro cent. (b) Supercontinuum white light source Fianium, NDF: neutral density filter, DM: dichroic mirror, L1: achromatic doublet (AC080-010-A-ML, $f = 10$ mm), L2: achromatic doublet ($f = 50$ mm), M: mirror, we: integrating sphere, S: spectrometer, P: prism spectrometer ($f_{\#} = 4.6$).

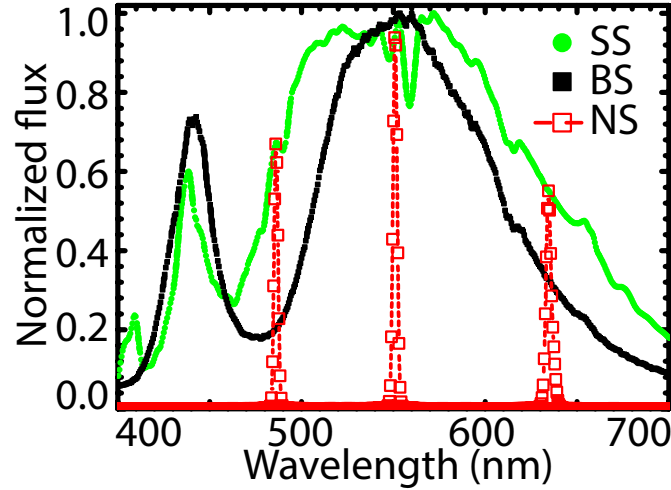


Figure 3.3: **Normalized reference spectra of the light sources used in the experiment.** Green circles - normalized reference spectrum of the supercontinuum source (SS) after being filtered by a neutral density filter and a dichroic mirror that removes the infrared part of the spectra. Black squares - normalized reference spectrum of a broadband source (BS) that was not filtered. Red squares - spectrally filtered narrowband light source (NS) normalized to the initial BS spectrum represent. We show narrowband spectra for three incident wavelengths $\lambda_i = 486, 551, 634$ nm. flux normalized to the SS.

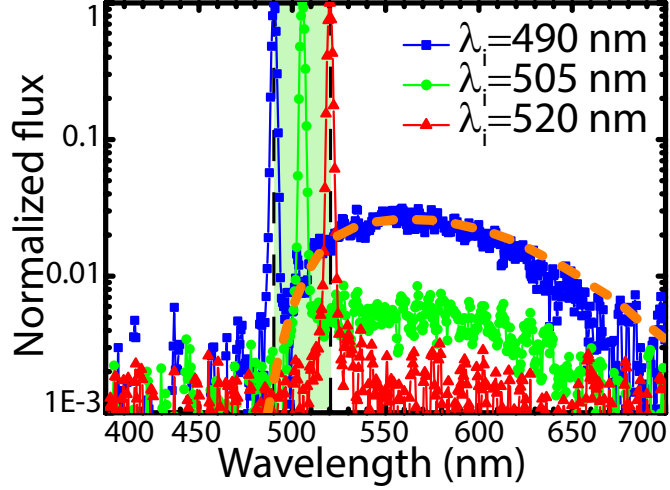


Figure 3.4: **Flux profile of the signal that we measure in the range where emission and absorption overlap for three different pump wavelengths.** The orange dashed line is the emission spectra from Fig. 3.1(a). The blue curve was normalized to 7791 counts and the green and the red curves to 15480 and 20297 counts respectively.

band and the broadband light source to check the consistency in the spectral ranges where both methods can be applied ($\lambda \leq \lambda_\ell$ and $\lambda \geq \lambda_r$). For the broadband light source, the scattered transmission is obtained by normalizing the measured spectrum F_T to a reference spectrum F_0 measured in the absence of a sample. For the narrowband light source, the scattered transmission is determined as the ratio of the transmitted flux $F_T(\lambda_i)$ and a reference flux $F_0(\lambda_i)$ without the sample at the designated wavelength λ_i that is set with the monochromator. The scattered transmission is reproducible to within a few percent points on different measurements with different light sources.

3.4 Results

3.4.1 Transmission measurements

To measure the scattered transmission $T(\lambda_i)$ of the polymer plates we tune the narrowband light source to an incident wavelength λ_i and measure the transmitted flux of the outgoing light. In Fig. 3.4 we show the normalized transmitted flux for three incident wavelengths λ_i . For an incident wavelength $\lambda_i = 490$ nm, we see a pronounced peak with the maximum at $\lambda = 490$ nm. This peak contains mostly elastically scattered photons because inelastically scattered photons are Stokes-shifted to longer wavelengths. Indeed between 500 nm and 650 nm, the flux profile reveals a broad peak that represents the Stokes-shifted flux since the flux profile has a shape similar to that of the YAG:Ce³⁺ in powder form (black dashed

curve). Both emission spectra have a maximum at 557 nm. For YAG:Ce³⁺ in powder form the flux is slightly higher at longer wavelengths $\lambda > 600$ nm than for the phosphor plate. The peak value of the Stokes-shifted light amounts to 3 % of the transmitted flux at the incident wavelength $\lambda_i=490$ nm. We calculate the amount of Stokes-shifted light F_{em} in the elastic peak at $\lambda_i = 490$ nm by extrapolating the emission curve to this wavelength range. We find that F_{em} amounts to less than 1 % of the transmitted flux F_{tot} , and can be safely neglected. The total Stokes-shifted flux decreases drastically for longer incident wavelengths λ_i inside the overlap range, and can thus be ignored too. For incident light in the middle of the overlap range at $\lambda_i = 505$ nm, we see that the emitted flux is even weaker with a normalized flux F_{em} less than 1% of F_{tot} . At the edge of the absorption band at the incoming wavelength $\lambda_i = 520$ nm we observe an even lower Stokes-shifted flux F_{em} . The reason for this decrease is that the absorption cross section decreases drastically in the overlap range, and only very little light is being absorbed, and as a result is Stokes-shifted. Throughout the overlap range, the contribution from the Stokes-shifted light F_{em} is less than 1 % of F_{tot} at the incident wavelength λ_i . We thus conclude that the Stokes-shifted flux contribution can be neglected throughout the overlap range. Therefore, we have distinguished the elastically scattered (or absorbed) fraction of light from the Stokes-shifted light. The amount of Stokes-shifted light strongly depends on the incident wavelength λ_i . As a result, we can now measure scattered transmission at any desired wavelength.

We have scanned the incident wavelength λ_i through the wavelength range of interest (see Fig. 3.4). In Fig. 3.1(b) the scattered transmission $T(\lambda)$ for the slab with 4 wt% of YAG:Ce³⁺ has been obtained from these scans (red squares). The total relative transmission $T_{\text{tot}}^{\text{rel}}(\lambda)$ measured with the broadband source is also shown in Fig. 3.1(b) for comparison (blue triangles). For short wavelengths, both transmission spectra coincide within a few percent. The spectrum measured with broadband light source reveals a deep trough with a minimum at 458 nm. The trough matches well with the peak of the absorption band of YAG:Ce³⁺ in Fig. 3.1(a), and reveals that the phosphor absorbs a significant fraction of the light in this wavelength range. At wavelengths longer than $\lambda_r = 520$ nm both transmission spectra are flat, but the spectrum measured with the broadband light source is 10 percent point larger than the spectrum measured with the narrowband light source. The spectrum measured with broadband light source contains a significant contribution of the Stokes-shifted light F_{em} in this spectral range, as most of the emission occurs in this spectral range (see Fig. 3.1(a)). The narrowband spectrum, on the contrary, does not have this contribution. The difference between these two spectra at long wavelengths is equal to $T_{\text{em}}(\lambda_i)$, in Eq. (3.2). In the overlap region ($\lambda_\ell = 490$ nm $< \lambda < \lambda_r = 520$ nm) both

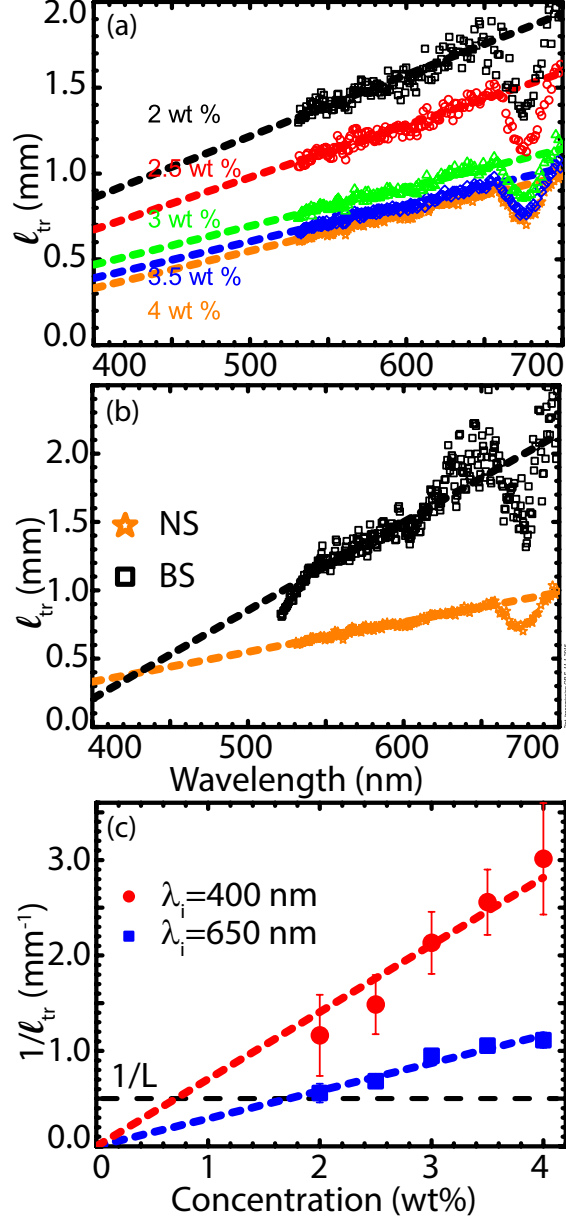


Figure 3.5: **Transport mean free path.** (a) Transport mean free path as a function of wavelength for different phosphor concentrations. Dashed lines represent linear fits to the measured data with parameters listed in table. 3.1. (b) Black squares indicate transport mean free path as a function of wavelength for broadband light source measurement. Orange stars indicate transport mean free path as a function of wavelength for narrowband light source measurement. Dashed lines represent linear fits to the measured data with parameters listed in table 3.1. Here concentration of the phosphor is taken to be 4 wt%. (c) Inverse transport mean free path as a function of concentration for two different incoming wavelengths. Dashed lines are linear fits to the data.

spectra reveal a sharp rise. In this range, the Stokes-shifted light causes the total relative transmission T_{tot}^{rel} to increasingly deviate from the scattered transmission. In Ref. [54] it

was not possible to separate elastically scattered and Stokes-shifted light.

Finally, we filtered the broadband light source with a long pass filter at $\lambda = 520$ nm, which ensures that the phosphor is not excited (we thus have zero emitted flux $T_{\text{em}}(\lambda_i)=0$). Hence, the measured total relative transmission $T_{\text{tot}}^{\text{rel}}$ equals the scattered transmission $T_{\text{tot}}^{\text{rel}} = T$ in this spectral range. In Fig. 3.1(b) we compare the scattered transmission measured with the filtered broadband light source (green), and the scattered transmission measured with the narrowband light source (red). The scattered transmission measured with the narrowband light source agrees within a few percent points with the scattered transmission measured with the filtered broadband light source (see Fig. 3.1(b)) for $\lambda \geq \lambda_r$. Concluding, we have for the first time extracted the scattered transmission $T(\lambda_i)$ for a diffuser plate with phosphor in the whole visible range, including the previously inaccessible overlap range.

3.4.2 Transport mean free path

In the range of low absorption ($520 \text{ nm} < \lambda < 700 \text{ nm}$) we have extracted the transport mean free path from the transmission data using Eq. 3.6 and plotted the result in Fig. 3.5(a). We see that the transport mean free path increases linearly with wavelength at constant phosphor concentration. In highly polydisperse non-absorbing media, a similar relationship between the transport mean free path and wavelength was found and successfully interpreted [53]. Therefore, we have fitted the transport mean free path with a line for every phosphor concentration. Parameters of the linear fits are listed in Table 3.1. We linearly extrapolate ℓ_{tr} to the absorption range $\lambda \leq \lambda_r = 520 \text{ nm}$, and use the extrapolated values of the transport mean free path ℓ_{tr} to obtain the absorption mean free path ℓ_a in the range of strong absorption. We investigated a systematic error in the transmission when the broadband light is used. We compare the apparent mean free path derived from $T_{\text{tot}}^{\text{rel}}$ with the true mean free path derived from T (Fig. 3.5(b)). The apparent mean free path clearly differs from the true one. The difference is nearly 100% in the range between 500 and 700 nm. Moreover, the slope is much steeper than for the true mean free path, as can be seen from Table 3.1.

In the limit of low concentration, each scatterer can be treated independently. In this case $1/\ell_{\text{tr}}$ is proportional to the concentration of the scatterers [70]. Indeed in Fig. 3.5(c), we see that the transport mean free path increases inversely proportional to the increasing phosphor concentration at a fixed wavelength. We observe that with increasing wavelength, the scattering cross section increases similarly to what was obtained earlier [54].

C (wt%)	a	b
2.0	-0.57 $\pm$ 0.07	0.00358 $\pm$ 0.00001
2.5	-0.55 $\pm$ 0.05	0.00305 $\pm$ 0.00008
3.0	-0.43 $\pm$ 0.03	0.00225 $\pm$ 0.00055
3.5	-0.47 $\pm$ 0.02	0.00214 $\pm$ 0.00038
4.0	-0.54 $\pm$ 0.02	0.00218 $\pm$ 0.00038
4.0 (BS)	-2.38 $\pm$ 0.14	0.00640 $\pm$ 0.00023

Table 3.1: Parameters of the linear models of the transport mean free path versus wavelength: $\ell_{tr} = a + b\lambda$. The parameters a and b depend on the phosphor concentration ρ and are shown with their standard errors. 4 (BS) - linear fit for data measured with the broadband light source.

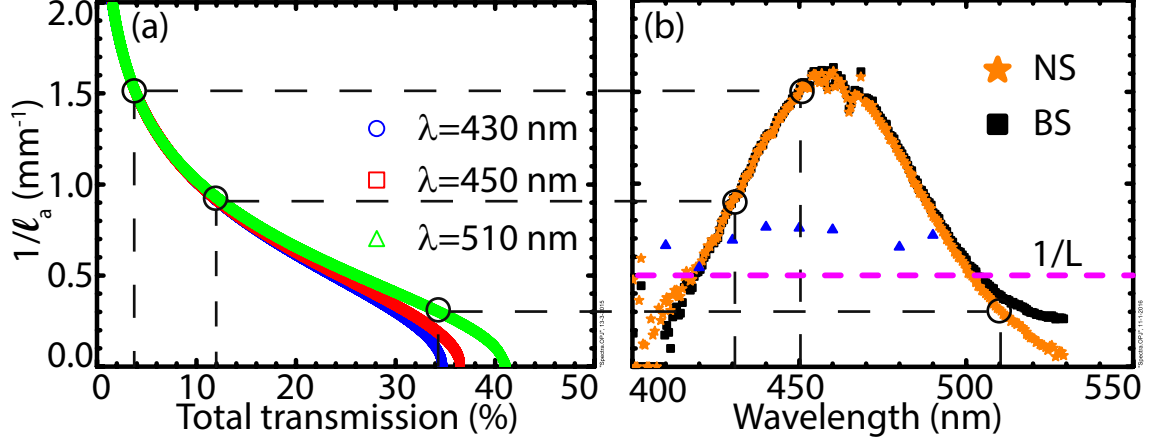


Figure 3.6: **Determination of the absorption mean free path.** (a) Look-up tables presented in a form of plots for Eq. 3.3 for three different wavelengths, and 4 wt% concentration of the phosphor. ($\ell_{tr} = 0.40; 0.44; 0.57$ mm respectively) (b) Orange stars show inverse absorption mean free path extracted from the look-up tables for the plate with 4 wt% phosphor concentration (narrowband light source). Black squares show inverse absorption mean free path extracted using broadband light source for the same plate. Black circles and black dashed lines indicate the process of mapping μ_a from the look-up table to the Fig. 3.6(b). Blue triangles indicate the inverse absorption length measured in Ref. [54]. The dashed purple line indicates the inverse thickness of the sample.

3.4.3 Absorption mean free path

By using the transport mean free path ℓ_{tr} extrapolated to the wavelength range between 400 and 530 nm where the phosphor absorbs light, we now derive the absorption mean free path ℓ_a from the measured scattered transmission (see Fig. 3.1(c)). Since we do not have an analytic inverse function of Eq. (3.3), we have solved the inversion numerically and made

look-up tables for each phosphor concentration and at each wavelength. Fig. 3.6(a) shows three inverted curves of μ_a versus scattered transmission for three different wavelengths ($\lambda = 430, 450, 510$ nm) at a phosphor concentration $\rho = 4$ wt%. We plot absorption coefficient μ_a as a function of transmission T because ℓ_a tends to infinity for vanishing absorption. Fig. 3.6(a) shows that μ_a (and thus the absorption) increase. In the limit of strong absorption, all scattered transmission curves tend to zero. In the limit of vanishing μ_a , the scattered transmission equals the scattered transmission from Eq. 3.6 that decrease with decreasing wavelength (Fig. 3.6(a)). The vertical dashed lines indicate the measured scattered transmission, and the intersections with the curves yield the corresponding μ_a for each wavelength at this phosphor concentration.

Fig. 3.6(b) shows the extracted absorption profile μ_a for the polymer plate with the highest phosphor concentration $\rho = 4$ wt% studied here. The FWHM of this curve is 64.5 nm. The absorption mean free path ℓ_a is shorter than the thickness of the sample d between 418 and 501 nm. This means that incident light is effectively absorbed in the volume of the sample, and the density of the phosphor is optimized for use in a white-light LED. At the edges of the absorption range at 400 and 530 nm μ_a tends to zero, as expected from the known absorption (Fig. 3.1(a)). We note that our present μ_a values differ from previous results obtained on the same samples [54] (see Fig. 3.6(b)). The absorption mean free path varies significantly with a wavelength in our case. We notably attribute the difference to the use of incorrect use of diffusion equation in Ref. [54]. The spectral shape obtained at present is in much better agreement with the phosphor absorption spectra than previously.

In Fig. 3.6(b), we also show the apparent inverse absorption length that is derived from the total relative transmission $T_{\text{tot}}^{\text{rel}}$ (BS). In a significant part of the absorption range ($420 < \lambda < 500$ nm), there is little difference in the exact absorption length derived from the scattered transmission T (NS). The reason for the small difference is apparent from Fig. 3.6(a). In Fig. 3.6(a) the absorption length differs little with the scattered transmission T for $T < 20\%$ independent of the transport mean free path. At low absorption ($\lambda < 500$ nm and $\lambda > 520$ nm) the difference between the apparent and the actual absorption mean free path becomes apparent.

In Fig. 3.7(a) we have plotted $\mu_a(\lambda)$ for three wavelength $\lambda = 430, 460$ and 500 nm as a function of phosphor concentration. We see that $\mu_a(\lambda)$ increases linearly with increasing phosphor concentration, which agrees with the assumption that each absorber is an independent absorber. Figure 3.7(a) shows that the steepest slope appears at the wavelength $\lambda = 460$ nm, which corresponds to the peak of the absorption curve. The maximum absorp-

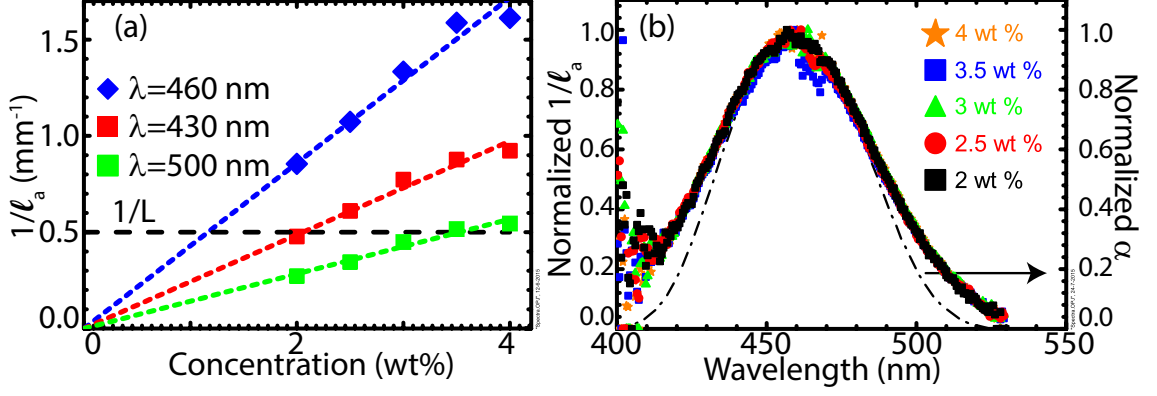


Figure 3.7: **Absorption mean free path.** (a) μ_a as a function of concentration is shown for the three different wavelength. (b) Line shapes of the absorption coefficient are shown for the phosphor in powder (dot-dashed black line) and in polymer plates with increasing phosphor concentrations.

tion cross section is $\sigma_a = 30 \mu m^2$, which agrees reasonably well with the typically measured absorption cross section for $YAG:Ce^{3+}$ of the order of $10 \mu m^2$ (Ref. [17, 71–75]).

Figure 3.7(b) shows the extracted normalized absorption spectrum of μ_a for the polymer plates with different phosphor concentrations. All absorption curves are normalized to their maximum, and they coincide with each other within a few percent points. The inverse absorption mean free path μ_a scales linearly with the phosphor concentration for all wavelengths. All absorption curves tend to zero outside $400 < \lambda < 530$ nm.

In Fig. 3.7(b) we also compare the shape of the absorption curve of the phosphor in powder form that was used to manufacture the samples to μ_a curves. The black dot-dashed line shows absorption spectra of the $YAG:Ce^{3+}$ in powder. These two sets of data were normalized to their maxima, so the positions of maximums of these two graphs coincide. The μ_a spectrum appear to have broader tails compared to the $YAG:Ce^{3+}$ in powder form. The absorption spectrum of $YAG:Ce^{3+}$ in powder has an FWHM = 54 nm that is 10 nm smaller than the FWHM of the measured absorption spectra. One possible reason for the difference is that light is multiply internally reflected in the $YAG:Ce^{3+}$ particles [17], so particles cannot be treated as independent scatterers.

Finally, let us place our approach in the context of previous work: Vos *et al.* reported the transport properties measurements in TiO_2 scattering plates using a broadband light source [53]. They showed that the transport mean free path ℓ_{tr} linearly depends on the wavelength in the visible wavelength range. This method cannot be applied to phosphor doped plates, as these plates have ranges with strong absorption, emission, and an overlapping range. Leung *et al.* reported the transport properties measurements in phosphor

doped slabs using a filtered broadband light source, where the linear dependence of ℓ_{tr} was exploited to calculate ℓ_{a} [54]. Unfortunately, however, the approximation used to analyze scattered transmission $T(\lambda_i)$ from Ref. [54] was used outside its range of validity. Secondly, the described method is limited to the region of strong absorption or emission but is not valid in the overlap region.

3.5 Summary and outlook

In this chapter, we have been able to separate light of the same wavelength yet originated from different physical processes occurring in the polymer plates of solid-state lighting units. This became possible by introducing narrowband light source in the setup. The separate measurement of elastically scattered and Stokes-shifted light allows us to extract the transport and absorption mean free path of the given polymer plates, which are the essential parameters required for modeling and predicting the color spectra of solid-state lighting devices. The optimal parameters of the solid-state lighting units can be directly extracted from the measured absorption curves. We vary the thickness of the plate d or the phosphor concentration depending on the desired level of pump absorption using absorption curve in Fig. 3.6(b).

We have developed a new technique to measure the light transport of white-light LED plates in the visible range based on narrowband illumination and spectrally sensitive detection. We compare the data obtained with the new technique to the data measured with the broadband light source. The two sets of data coincide in the range without absorption $\lambda \geq \lambda_r$, as well as in the range with strong absorption $\lambda \leq \lambda_\ell$. We extracted the scattered transmission in the overlap range that was previously inaccessible.

We used diffusion theory to extract transport ℓ_{tr} and absorption ℓ_{a} mean free paths from these data in the previously inaccessible range. The shape of the absorption coefficient measured for the YAG:Ce³⁺ powder, and YAG:Ce³⁺ powder in a polymer matrix have similar trends, although for the polymer plates the curve is broader than for the YAG:Ce³⁺ powder. Both μ_{a} and $1/\ell_{\text{tr}}$ are proportional to the concentration of phosphor, which reveals that elastic and inelastic processes do not influence each other.

By exploiting narrowband light source and interpreting the resulting scattered transmission by diffusion theory, we extracted for the first time light transport parameters for white LEDs in the whole visible wavelength range. However, diffusion theory only gives an analytical solution for simple sample geometries, such as a slab, a sphere, or a semi-infinite medium. Therefore, to efficiently model a real white LED with a complex geometry

we must, in the end, supplement an *ab initio* theory with a numerical method. An important class of numerical methods is the Monte Carlo methods, wherein the case of light in arbitrary geometries the ray tracing techniques are a major workhorse [15–18]. Future work should, therefore, focus on the question of how to combine analytical methods with numerical ones.

Chapter 4

Accurate determination of transport parameters in strongly absorbing and scattering materials

4.1 Introduction

Currently, light propagation through scattering and strongly absorbing optical systems, like white LEDs, is described using various numerical methods such as Monte Carlo simulations and ray tracing [15–18, 76]. While the computational methods incorporate the detailed geometry of the problem at hand, they also have several substantial limitations. There is a trade-off between the obtained precision and the speed of numerical approaches that ultimately compromises the design of white LEDs, since accuracy increases with the number of numerical samples taken, thereby naturally decreasing speed. Moreover, the accuracy of numerical methods is not always beyond doubt, as *a posteriori* readjustments of the transport parameters have been reported [17, 18]. Numerical methods do not possess predictive power for different LEDs than the one calculated, as opposed to analytical methods. Hence for every new situation (*e.g.*, new phosphor, new blue LED, etc.) new simulations must be performed. Therefore, it is timely to search for analytic approaches that have predictive power (even outside the immediately studied parameter domain), and that are accurate and fast.

In the optically thin samples $d/\ell_{\text{tr}} \ll 1$ (see Fig. 4.1), the transport of light is analytically described by the venerable Lambert-Beer (or Beer-Lambert-Bouguer) law. In scattering systems such as white LEDs, however, the condition $d/\ell_{\text{tr}} \approx 1$ holds [53, 54], hence light is scattered multiple times and diffused ¹. Moreover, we will see that in certain

¹To be sure, in a scattering system the degree of scattering is given by d/ℓ_s rather than d/ℓ_{tr} . Since in

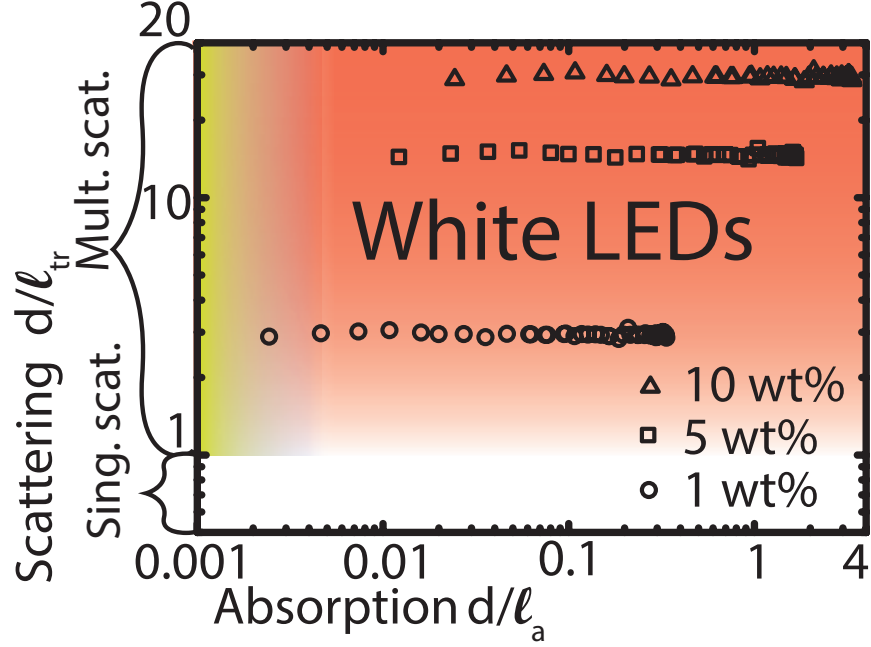


Figure 4.1: **Transport parameters in the plane spanned by absorption and diffusion.** The absorption on the abscissa is gauged by the ratio of sample thickness d and absorption mean free path ℓ_a and the diffusion strength on the ordinate by the ratio of sample thickness d and transport mean free path ℓ_{tr} . The shaded yellow range indicates the area where the diffusion theory performs well. The red range represents the transport parameter range accessed in the current manuscript. Symbols represent presently measured absorption and scattering as a function of wavelength for phosphor diffuser plates with particle concentrations 1 – 10 wt%. The gradient on the figure shows artistic representation of the diffusion theory applicability.

common circumstances, light is also strongly absorbed $d/\ell_a \geq 1$. A widely known theory of transport of light is the radiative transfer equation [42]. While the full radiative transfer equation requires numerical solutions (for example Monte Carlo simulations), there are several relevant cases where analytical solutions exist. The first order approximation to radiative transfer equation is the diffusion equation (see Ch. 2) [23, 25, 26, 77, 78]. Solutions to the diffusion equation work surprisingly well even in regimes where they are not supposed to function [23] such as optically thin samples [24, 53]. Regarding absorption, however, the validity of the diffusion theory is limited to weakly absorbing systems with $d/\ell_a \ll 1$, as indicated in Fig. 4.1 and Ch. 6 [43, 77]. In white LEDs, however, the absorption mean free path at maximum absorption is at least four times smaller than the sample thickness

the vast majority of scattering systems the transport mean free paths exceeds the scattering mean free path ($\ell_{tr} \geq \ell_s$) due to predominantly forward scattering, we consider our characterization to be on the safe side.

$d/\ell_a \geq 4$, see Fig. 4.1. Since in this regime the diffusion theory fails to describe the light propagation, other analytical approaches are required to accurately determine transport parameters of scattering materials with absorption, and to offer fast design tools for white LEDs.

In this chapter, we describe an analytical, accurate, and fast approach to determine the transport parameters in samples that are used in white LEDs. The determination of the transport parameters involves the measurement of the total transmission, the scattered reflection, and the ballistic transmission for a set of diffuser plates as a function of the concentration of the phosphor particles. The total transmission and the scattered reflection data are analyzed using the P3 approximation to the radiative transfer equation (RTE) [33]. In optically thin scattering samples where the contribution of the ballistic light dominates, the data were analyzed using the Lambert-Beer law. To verify our approach, we performed Monte Carlo simulations and found excellent agreement between the numerically obtained transport parameters and analytically obtained transport parameters. The demonstrated approach offers a fast and accurate determination of the transport parameters and possesses predictive power beyond the parameter range studied. The precision of the extracted transport parameters is only limited by the errors of the experimental data. Besides, the proposed approach of measuring the total transmission T and the scattered reflection R allows us to decouple the scattering and the absorption in a straightforward experimental way. Once transport parameters are known, they can be used as input parameters in the ray tracing Monte Carlo calculations for white LED design which requires complex geometries that are not captured by the analytical approach [79]. We show that the parameter extraction is also applicable within the domain of validity of the diffusion theory (see Ch. 6). Hence, the proposed parameter extraction approach pertains also to the propagation of light in other relevant application fields, such as oceans and clouds [80–84], pharmaceutical products [38–40], and in noninvasive diagnostic imaging of living tissues [32–37].

4.2 Model

In this chapter the diffuser plate with phosphor particles is modeled as a slab of thickness d that extends from $z = 0$ to $z = d$, as shown in Fig. 4.2. Incident plane waves at wavelength λ with flux $F_i(\lambda, z = 0, \rho) = F_i(\lambda, 0, \rho)$ illuminate the slab, where ρ is the particle density. The incident light is scattered multiple times and possibly absorbed inside the slab. We characterize the light scattering and absorption in the slab by measuring the total transmission T , the scattered reflection R , and the ballistic transmission T_b that are

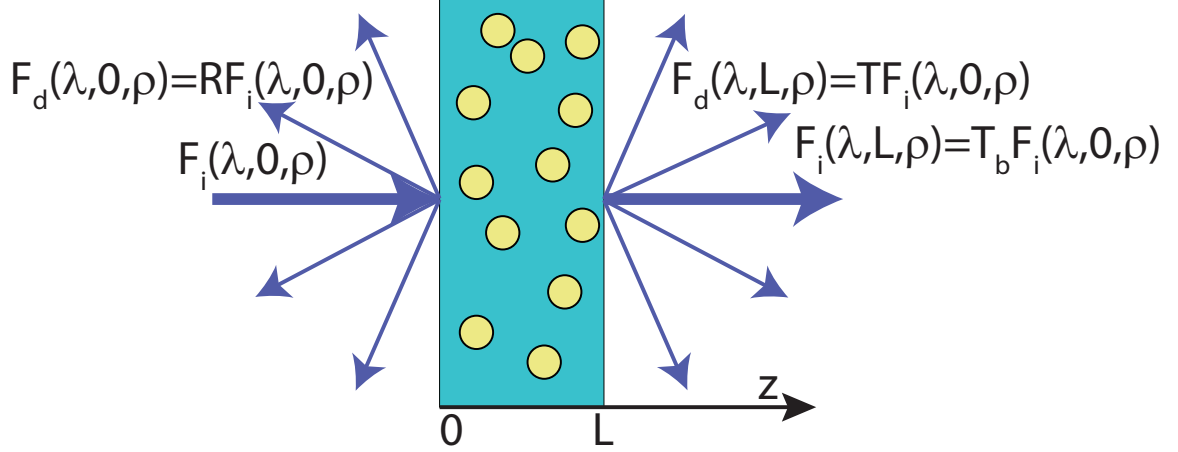


Figure 4.2: **Scheme of light incident on and exiting from a slab.** Plane waves with flux $F_i(\lambda, 0, \rho)$ are incident in the z -direction on a slab of scattering material that contains phosphor particles (represented by yellow circles). $F_d(\lambda, 0, \rho)$ is the diffuse reflected flux, $F_d(\lambda, d, \rho)$ is the diffuse transmitted flux, and $F_i(\lambda, d, \rho)$ is the transmitted ballistic flux. T and R are the total transmission and the scattered reflection, respectively, and T_b is the ballistic transmission.

defined as

$$T(\lambda, d, \rho) \equiv \frac{F_T(\lambda)}{F_0(\lambda)} = \frac{F_d(\lambda, d, \rho) + F_i(\lambda, d, \rho)}{F_i(\lambda, 0, \rho)}, \quad (4.1)$$

and

$$R(\lambda, 0, \rho) \equiv \frac{F_d(\lambda, 0, \rho)}{F_i(\lambda, 0, \rho)}, \quad (4.2)$$

and

$$T_b(\lambda, d) \equiv \frac{F_i(\lambda, d, \rho)}{F_i(\lambda, 0, \rho)}, \quad (4.3)$$

where $F_T(\lambda)$ is the elastic transmitted flux, $F_d(\lambda, 0, \rho)$ and $F_d(\lambda, d, \rho)$ are the diffused flux integrated over all outgoing angles at the positions $z = 0$ and $z = d$, respectively, and $F_i(\lambda, d, \rho)$ is the ballistic transmitted flux. The measured total transmission T , the scattered reflection R , and the ballistic transmission T_b explicitly depend on the experimentally available parameters: the wavelength λ , the thickness of the sample d , and the density ρ of the phosphor particles. We will use measured total transmission T , scattered reflection R , and ballistic transmission T_b to extract average single particle properties: the scattering cross section σ_s , the absorption cross section σ_a , and the anisotropy factor μ [41].

To extract the average single particle properties $(\sigma_s, \sigma_a, \mu)$ we exploit an analytical model to compute the total transmission T^m , the scattered reflection R^m and the ballistic transmission T_b^m that not only explicitly depend on the wavelength λ , the thickness of the sample d , and the density ρ of the phosphor particles, but also on single particle properties

$(\sigma_s, \sigma_a, \mu)$. By equating (T^m, R^m, T_b^m) to the experimentally measured (T, R, T_b) we compute the transport parameters $(\sigma_s, \sigma_a, \mu)$. As the analytical model we chose to employ the P3 approximation to the radiative transfer equation [33, 35–37, 45, 51, 85]

$$T^m(\lambda, d, \rho; \sigma_s, \sigma_a, \mu) \equiv \frac{F_d(\lambda, d, \rho; \sigma_s, \sigma_a, \mu) + F_i(\lambda, d, \rho; \sigma_s, \sigma_a, \mu)}{F_i(\lambda, d, \rho; \sigma_s, \sigma_a, \mu)}, \quad (4.4)$$

and

$$R^m(\lambda, d, \rho; \sigma_s, \sigma_a, \mu) \equiv \frac{F_d(\lambda, d, \rho; \sigma_s, \sigma_a, \mu)}{F_i(\lambda, d, \rho; \sigma_s, \sigma_a, \mu)}, \quad (4.5)$$

with

$$F_d(\lambda, z, \rho; \sigma_s, \sigma_a, \mu) = \sum_{i=1}^4 B_{1i} \exp(\mu_i z) + G_1 \exp(-\rho \sigma_t z), \quad (4.6)$$

where $\sigma_t = \sigma_s + \sigma_a$ is the sum of the scattering cross section σ_s and the absorption cross section σ_a , and B_{mi} when $m = 1$, G_m when $m = 1$ and μ_i are functions described in the Ch. 2, ρ is the particle concentration in the samples, and $F_i(\lambda, d, \rho; \sigma_s, \sigma_a, \mu)$ is the incident light flux and $I_d(\lambda, z, \rho; \sigma_s, \sigma_a, \mu)$ the flux of the light at position z .

The ballistic flux that is transmitted through the sample is described by the Lambert-Beer law

$$T_b^m(\lambda, d, \rho; \sigma_a, \sigma_s) \equiv \frac{F_d(\lambda, z, \rho; \sigma_s, \sigma_a, \mu) e^{-(\rho \sigma_t d)}}{F_d(\lambda, z, \rho; \sigma_s, \sigma_a, \mu)} = \exp(-\rho \sigma_t d). \quad (4.7)$$

Knowledge of the average single particle scattering properties $(\sigma_s, \sigma_a, \mu)$ at any wavelength λ allows us to *predict* the light transport properties for any scattering and absorbing sample for any particle density ρ and sample thickness d outside the immediately studied density and thickness range. Therefore, it is convenient to express the single-particle properties into the scattering mean free path ℓ_s the transport mean free path ℓ_{tr} and the absorption mean free path ℓ_a from the following relationships [54, 86]

$$\ell_s = \frac{1}{\rho \sigma_s}, \quad (4.8)$$

$$\ell_a = \frac{1}{\rho \sigma_a}, \quad (4.9)$$

$$\frac{1}{\ell_{tr}} = \frac{(1 - \mu)}{\ell_s} + \frac{1}{\ell_a}, \quad (4.10)$$

provided the underlying assumptions of the independent scatterer approach [23] namely negligible multiparticle effects, negligible re-absorption, and negligible absorption of the

medium in which particles are embedded remain fulfilled.

4.3 Experimental details

The samples studied in the experiments are $d = 1.98 \pm 0.02$ mm thick silicone plates (polydimethylsiloxane, PDMS, refractive index $n = 1.4$ [87]) that are doped with YAG:Ce³⁺ phosphor particles in a range of particle concentration ρ ranging from 1 to 10 weight percent phosphor. The absorption spectrum of the phosphor particles has a peak absorption wavelength $\lambda = 458$ nm, and a broad linewidth (full width half maximum, FWHM) of $\Delta\lambda_a = 55$ nm, see Ch. 3. Based on the absorption spectrum we choose the wavelength $\lambda_r = 520$ nm to distinguish the absorbing range ($\lambda = 400$ nm to 520 nm) from the non-absorbing range ($\lambda = 520$ nm to 700 nm), as in Ch. 3.

Figure 4.3 shows a schematic of the experimental setup used to measure the total transmission T , the total reflectance R and the ballistic transmission T_b . The sample is illuminated with a tunable narrowband light source in the visible wavelength range 420 – 700 nm. To this end, the beam of a supercontinuum white-light source (Fianium WL-SC-UV-3) is spectrally filtered to an adjustable bandwidth $\Delta\lambda < 2.4$ nm using a prism monochromator (Carl-Leiss Berlin-Steglitz). Unwanted emission of the supercontinuum source in the infrared $\lambda_i > 700$ nm is blocked with a neutral density filter (NENIR30A) and a dichroic mirror (DMSP805), see Fig. 4.3(a).

The incident beam illuminates the sample at normal incidence. The scattered flux is collected using either an integrating sphere or a ballistic detector. The integrating sphere (Opsira uku-240) has three entrance ports, each with a 20 mm diameter. Each port can be selectively closed with a baffle that has the same diffusive inner coating as the remainder of the integrating sphere. The entrance port of the integrating sphere is sufficiently large to collect scattered light that emanates at all angles from the strongest scattering sample. The flux of the outgoing light entering the integrating sphere is analyzed with a fiber-to-chip spectrometer (AvaSpec-USB2-ULS2048L) with a spectral resolution $\Delta\lambda_s = 2.4$ nm.

To measure the diffuse reflectance R , the ports 1 and 3 of the integrating sphere are used, see Fig. 4.3(b). The sample is attached to port 3 with the incident light flux $F_i(\lambda, 0, \rho)$ entering the sphere through port 1. The diffuse reflected light flux $F_d(\lambda, 0, \rho)$ is collected with the fiber spectrometer. The reference flux $F_i(\lambda, 0, \rho)$ separately enters the sphere under an angle through port 1, in such way that the beam is incident on the surface of the sphere but not onto the sample.

To measure the total transmission, the ports 1 and 2 are used, as shown in Fig. 4.3(c).

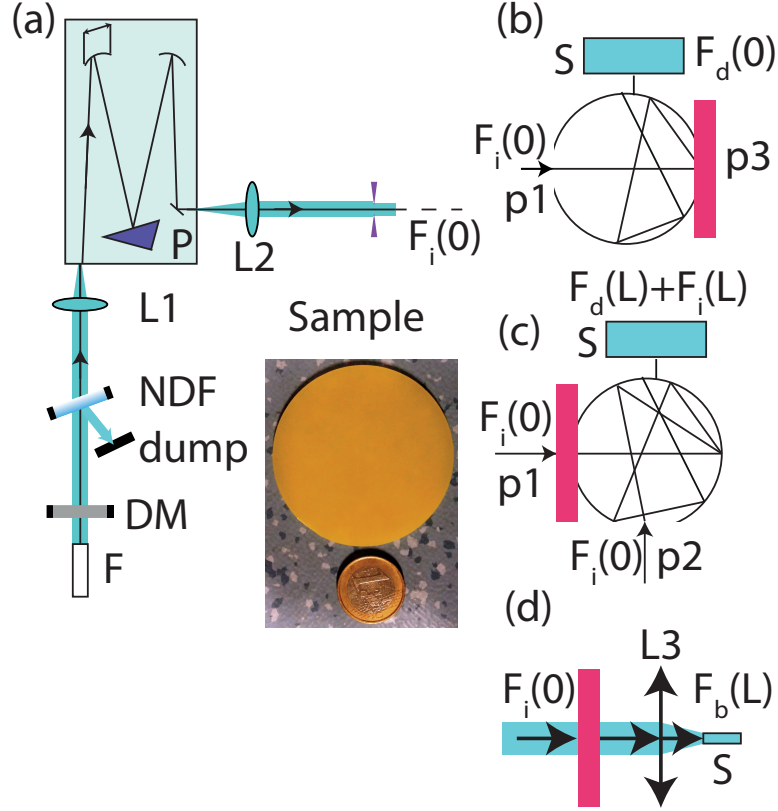


Figure 4.3: **Experimental setup for measuring transport parameters.** (a) The light source used in the experiment. (b) The configuration of the integrating sphere used for the scattered reflection measurements. (c) The configuration of the integrating sphere used for the total transmission measurements. (d) The configuration for ballistic Lamber-Beer measurements. F: Supercontinuum white light source, NDF: Neutral density filter, DM: Dichroic mirror, L1: Achromatic doublet (AC080-010-A-ML, $f = 10$ mm), L2: Achromatic doublet ($f = 50$ mm), L3: Achromatic doublet (AC254-050-A-ML, $f = 50$ mm), I: Integrating sphere, S: Spectrometer, P: Prism monochromator ($f_{\#} = 4.6$), p1: port1, p2: port 2, p3: port 3.

The sample is attached to port 1 with the incident light flux $F_i(\lambda, 0, \rho)$ illuminating the sample. The transmitted light is collected by the integrating sphere and spectrally resolved with the fiber spectrometer. In the transmission configuration, the reference flux $F_i(\lambda, 0, \rho)$ is separately measured with the incident light entering sphere through port 2.

The ballistic detector measures the transmitted ballistic light T_b . The scheme of this detector is shown in Fig. 4.3(d). It consists of an achromatic lens ($f = 50$ mm) that collects the transmitted light into the entrance of a fiber spectrometer (AvaSpec-USB2-ULS2048L). The distance between the detector and the sample is set to 50 cm. If the detector is placed closer to the sample, the ballistic signal is compromised by a significant contribution from the diffuse scattered light. The contribution of the scattered light in the ballistic signal is

estimated following Ref. [88] to be less than 1%.

The measurements of the transmission T , the reflection R and the ballistic transmission T_b are repeated 10 times at every wavelength, to obtain statistical information. Consequently, we estimated the error to be about $\Delta T/T = \Delta R/R = 4$ percent point for transmission and reflection measurements, and about $\Delta T_b/T_b = 1$ percent point for the ballistic transmission measurements. In Fig. 4.5 and Fig. 4.6 the error bars are within the symbol size. The estimated errors of the total transmission T and the scattered reflection R are propagating linearly when the transport parameters are calculated, and result in a 4 percent point error on the extracted transport parameters $(\sigma_s, \sigma_a, \mu)$.

4.4 Simulations

We perform Monte Carlo ray tracing simulations, in other words, Monte Carlo simulations of the radiative transfer equation as independent verification of our analytical model. The Monte Carlo ray tracing was carried out using a computational geometry similar to the schematic in Fig. 4.2 with normally incident monochromatic plane wave on a plane parallel slab with finite thickness d . The scattering parameters (ℓ_s, ℓ_a, μ) for the chosen wavelength are taken as the input parameters for the simulations. The light transport is simulated as follows [89, 90]: A bunch of $N_b = 10^4$ random walkers (photons) is launched with initial angular coordinates $(\theta_0, \phi_0) = (0 \text{ rad}, 0 \text{ rad})$ representing the normally incident plane wave. The incident photon bunch undergoes a three-dimensional random walk inside the slab. The random walk consists of a set of connected rectilinear paths of lengths $\{d_i\}$, each of which is randomly picked from an exponential distribution $p(d_i) = (1/\ell_s) \exp(-d_i/\ell_s)$. The scattering at the end of each path is simulated by choosing an angular coordinate (θ_i, ϕ_i) following the Henyey-Greenstein phase function [89] to accommodate the scattering anisotropy μ of the scatterers. Absorption in the medium is incorporated through the exponential decrease of the photon number: the attenuation depends exponentially on the path length d_i and the absorption mean free path ℓ_a . The random walk is terminated when the photon bunch is either completely absorbed, or it has arrived at an interface. The specular light reflection from the interface is described with Fresnel's law [19]. The amount of transmitted photons N_{tr} , reflected photons N_{ref} , or absorbed photon N_{abs} weight is registered at the end of the walk. Simulated transmission is defined as $T^s \equiv N_{tr}/N_b$, and simulated reflection as $R^s \equiv N_{ref}/N_b$. A total of about $N_{ph} = 2 \times 10^4$ photon bunches are launched to statistically compute the transmission and reflection for a given set of transport parameters (ℓ_s, ℓ_a, μ) . The number of photons N_{ph} is chosen to ensure the convergence of

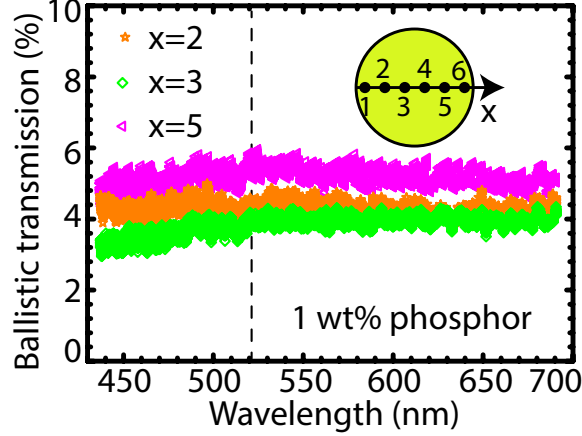


Figure 4.4: **Ballistic transmission of the phosphor slab obtained in the experiment.** of the silicone plate with 1 wt% of phosphor particles measured at different spatial positions. The beam position during the measurements is shown in the inset.

the algorithm within 1% of the transmission fluctuations between different algorithm runs. We scan the transport parameter range ($\mu \in [0, 1]$, $\sigma_s \in [1, 3]$ and $\sigma_a \in [0, 0.5]$) for different phosphor concentrations and choose the transport parameters that yield the transmission and the reflection obtained in the experiment.

4.5 Results

4.5.1 Ballistic transmission

The samples with 1 wt% and 2 wt% phosphor concentration were used for the ballistic transmission measurements since they are optically sufficiently thin. The ballistic transmission for other plates is less than 0.1%, which is below the signal to noise level of our detector at the available incident signal intensities.

We scan the incident wavelength λ_i from 420 nm to 700 nm, and collect the ballistic light, with light incident at six different spatial positions on the sample as shown in the inset of Fig. 4.4. Representative data sets are shown in Fig. 4.4, data for positions 1, 4, and 6 are in between the presented data sets. The ballistic transmission spectra for the slab with 1 wt% is shown at three selected spatial positions in Fig. 4.4. The ballistic transmission at wavelength λ_i differs at most by 2 percent points at different spatial locations $\Delta T_b/T_b < 2\%$. The difference between different data sets arises from the exponential sensitivity of the ballistic light to variations in the local phosphor particle concentration, see Eq. (4.7).

We extract the sum of the scattering and absorption cross sections $\sigma_s + \sigma_a$ from the measured ballistic transmission data using Eq. (4.7). The results are plotted in Fig. 4.5

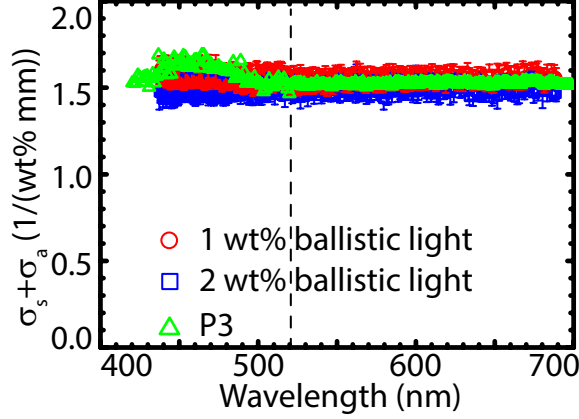


Figure 4.5: **Measured scattering and absorption cross sections.** The sum of the scattering cross section σ_s and absorption cross section σ_a measured as a function of wavelength for two different phosphor concentrations. Green triangles represent data extracted from the diffused light measurements.

for the samples with 1 wt% and 2 wt% concentrations of phosphor particles. We observe excellent agreement between samples with different concentrations of phosphor in the wavelength range of interest. In the non-absorbing range (520 nm to 700 nm), the extracted cross section is equal to the scattering cross section σ_s since the absorption cross section vanishes.

In the absorption range, the measured ballistic transmission is constant within the signal to noise ratio. From this behavior, we conclude that the absorption cross section σ_a is at least one order of magnitude smaller than the scattering cross section σ_s . In the case of the multiple scattering even this small absorption has a crucial influence on the diffuse transmission, and reflection (see subsequent sections).

4.5.2 Diffuse light transmission and reflection

We measure the transmission and the reflection spectra for the slabs with different phosphor particle concentrations. The measured results are shown in Fig. 4.6(a) and 4.6(b). For visual clarity, we plot only a selection of the measured samples. The transmission and the reflection spectra reveal a deep trough with a minimum at $\lambda_i = 458$ nm. The trough matches the peak of the absorption band of YAG:Ce³⁺ [86]. The presence of the trough indicates that the phosphor absorbs a significant fraction of the light in this wavelength range. In the range $\lambda_\ell = 490$ nm $<$ $\lambda_r <$ 520 nm where absorption and emission of phosphor overlap (see Ch. 3) [86], the spectra show a steep rise in the transmission due to the reduced absorption. At wavelengths longer than $\lambda_r = 520$ nm, the transmission spectra are flat. Unlike the transmission spectra, the scattered reflection spectra depend only weakly on the phosphor

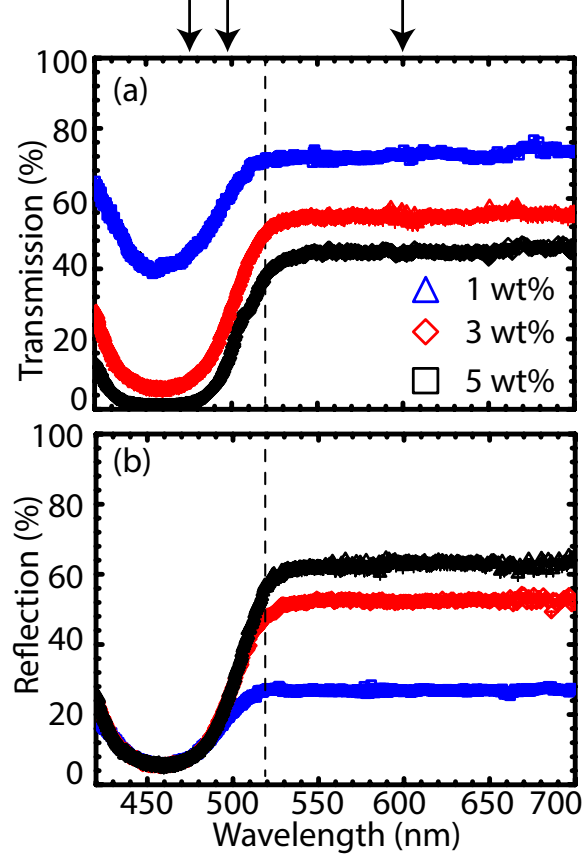


Figure 4.6: **Transmission and scattered reflection spectra of silicone plates.** (a) Broadband transmission T as a function of concentration. (b) Broadband reflection as a function of concentration. Concentration of the phosphor particles is indicated on the figures. Arrows indicate the wavelengths for the Fig. 4.7

concentration ρ in the absorption range up to $\lambda_\ell = 490$ nm Fig. 4.6(b).

The sum of the total transmission and the scattered reflection was found to be 104 % for all the measurements in the non-absorbing range. This small systematic error could arise from an overestimation of the diffuse light signal. A possible source of the excess signal is the specular light (see Fig.4.3). Approximately 2 % of the reference light is being specularly reflected (Fresnel reflection) from the sample and is lost in the setup. Part of the specularly reflected light can also illuminate the sample, however, resulting in an effectively higher flux that illuminates the sample.

We analyze experimental results by extracting the transport parameters using the P3 approximation model Eq. (4.4) and (4.5) to the total diffuse transmission and the total diffuse reflection data. Since the theoretical description requires energy conservation with the diffuse reflection and transmission adding to 100 %, we decided to fit T and R separately given the slight systematic error mentioned above. As an example, the fit at $\lambda_i = 599$ nm

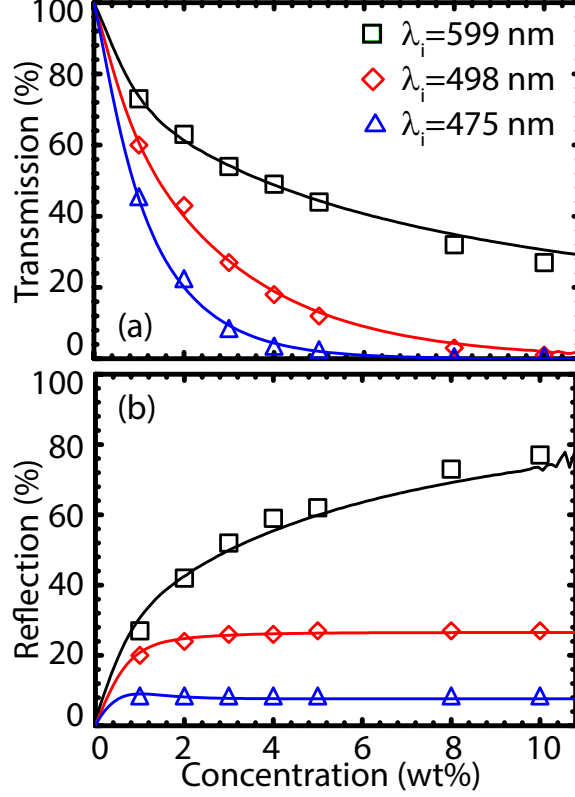


Figure 4.7: **Transmission and reflection as a function of concentration for three chosen wavelengths.** (a) Transmission as a function of concentration shown for three different wavelengths. (b) Reflection as a function of concentration shown for the same three different wavelengths. In both figures, symbols represent measured values. Solid lines are analytical results obtained using the P3 approximation to the radiative transfer equation.

is shown in Fig. 4.7. The difference between the anisotropy parameter $\mu(\lambda = 599 \text{ nm})$ extracted from T and R is about 4 % percent, which is within the error of the experiment. Fig. 4.7 illustrates the model prediction at three wavelengths in the range of strong absorption ($\lambda_i = 475 \text{ nm}$), weak absorption ($\lambda_i = 498 \text{ nm}$), and no absorption ($\lambda_i = 599 \text{ nm}$). The transmission is a monotonically decaying function of the concentration ρ over the studied wavelength range. The diffuse reflection is an increasing function of the concentration ρ in the wavelength range with no absorption. In the absorption range, the increase of the reflection R in the high concentration range is hampered by the absorption.

For each wavelength λ_i in the absorption range ($400 < \lambda_i < 490 \text{ nm}$), we fit the analytic (T^m , R^m) simultaneously to the measured (T, R) as a function of the concentration ρ , with the scattering cross section σ_s , the absorption cross section σ_a , and the anisotropy factor μ as the adjustable parameters. In the non-absorbing range, $490 \text{ nm} < \lambda_i < 700 \text{ nm}$ the scattering cross sections σ_s is taken from the ballistic light measurements. For the stability

of the procedure, we take the absorption cross section to be small $\sigma_a = 10^{-7}$, where we verified that even a decrease of σ_a by several orders of magnitude does not affect the result. The only adjustable parameter in the non-absorbing range is the anisotropy factor μ . If the total transmission has sharply decreased to $T < 1\%$, the analytical model reveals strong oscillations, as shown in Fig. 4.7(b). These oscillations are the result of finite machine precision in calculating the numerical values of the total transmission T , and the scattered reflection R using Eq. 4.4 and 4.5.

In Fig. 4.8 the extracted parameters (σ_s , σ_a , μ) are shown for the whole visible wavelength range. In the whole wavelength range the scattering cross section σ_s , and the anisotropy factor μ remain constant with the incident varying wavelength λ_i . In the absorption range, the maximum absorption cross section σ_a coincides with the absorption peak of the YAG:Ce³⁺. The absorption cross section tends to zero at the edges of the absorption spectral range ($\lambda_i = 420$ nm and 520 nm) in agreement with [86]. To compare the P3 approximation to the diffusion equation (P1 approximation), we determine the absorption cross section σ_a using both equations. The diffusion equation shows up to 30 % error in the absorption cross section σ_a , and hence cannot be used for strongly absorbing materials, see Fig. 4.8(c).

To verify the derivation of the parameters (σ_s , σ_a , μ) by our analytical model, we employ Monte Carlo simulations of the radiative transfer equation. At every wavelength λ , we simulate (T^s, R^s) as a function of the parameters (ℓ_s, ℓ_a, μ) to within 4 % of the measured data (T, R) . For a particular concentration ρ this comparison yields the parameters (ℓ_s, ℓ_a, μ) as a function of λ . We compute the parameter estimates for all measured concentrations ρ . We verify that (ℓ_s, ℓ_a) are proportional to the density ρ , and that μ remains fixed for all densities ρ . The $(\sigma_s, \sigma_a, \mu)$ at a given wavelength λ is the best fit parameter set for all concentrations. We convert (ℓ_s, ℓ_a) to (σ_s, σ_a) using Eq. (6.2) and Eq. (6.3).

The simulation results reveal good agreement with the analytical P3 results, as shown in Fig. 4.8, where the error bars in the Monte Carlo parameter estimate is a result of the regression analysis. Hence the transport parameters of white LED can be extracted equally well with the P3 approximation to the radiative transfer equation (RTE) as with the Monte Carlo simulations. We also compare the sum of the scattering cross section σ_s and the absorption cross section σ_a with the sum obtained from the ballistic light measurements in the absorption range also shown in Fig. 4.5. Both approaches yield a sum $(\sigma_s + \sigma_a)$ that is in excellent agreement. In the absorption range ($420 < \lambda < 520$ nm) the P3 approximation reveals a shallow peak that is not apparent in the ballistic data, most likely due to the limited signal to noise ratio in the latter. Thus, the transport parameters extracted with the P3

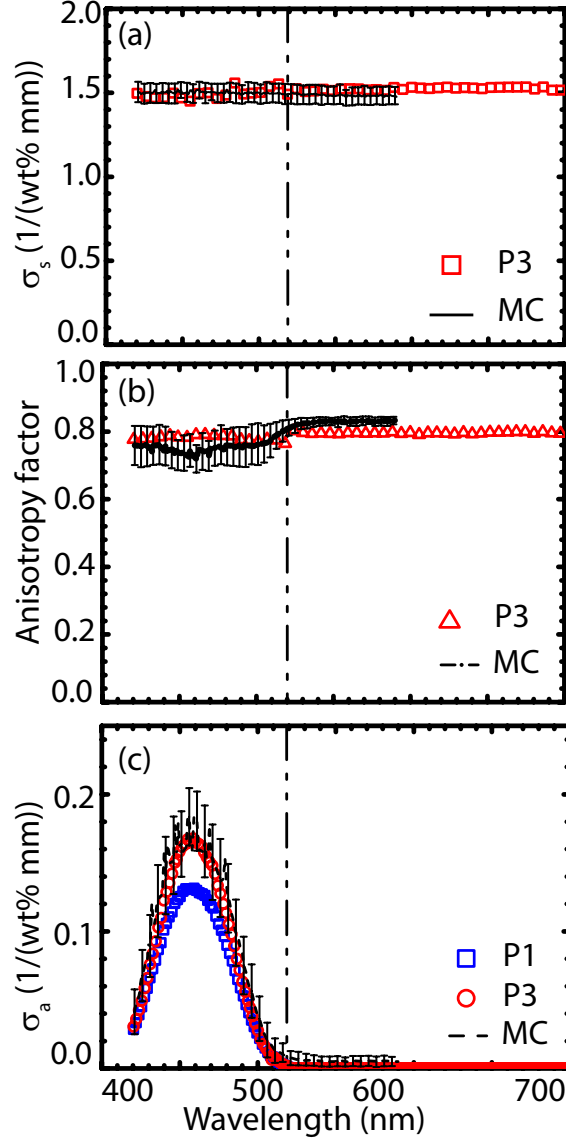


Figure 4.8: **Transport parameters of YAG:Ce³⁺ phosphor particles extracted using analytical model and Monte Carlo simulations.** Open symbols are the transport parameters values extracted using an analytical approach. The solid, dashed, dot-dashed lines are the Monte Carlo simulation results with respective error bars. **(a)** Scattering cross section σ_s as a function of wavelength. **(b)** Anisotropy factor μ as a function of wavelength. **(c)** Absorption cross section σ_a as a function of wavelength calculated using diffusion theory (P1), P3 approximation, and using Monte Carlo simulations.

approximation to the RTE are in good agreement with both the ballistic light determination and with the Monte Carlo estimations, which confirms the validity of our analytical model.

4.6 Discussion

We verified that our analytical method is superior to numerical methods in speed and precision: It takes ~ 1 min at each wavelength to obtain the transport parameters with error margins $\Delta\sigma_s/\sigma_s \leq 4\%$, $\Delta\sigma_a/\sigma_a \leq 4\%$ and $\Delta\mu/\mu \leq 4\%$ with the analytical approach. The extraction speed of the analytical method can be further greatly optimized using dedicated solvers. The precision of the analytical method within the much shorter processing time is only limited by the precision of the measured input data. To achieve comparable precision it takes ~ 17 min on conventional PC for a Monte Carlo approach with even larger error margins $\Delta\sigma_s/\sigma_s \leq 4\%$, $\Delta\sigma_a/\sigma_a \leq 10\%$ and $\Delta\mu/\mu \leq 7\%$. The precision of the Monte Carlo simulations improves only relatively slowly as it scales as the square root of the number of rays used. Another advantage of the analytical approach over numerical methods is the predictive power over a broad range of transport parameters where $d/\ell_a < 7$.

Let us place our approach in the context of previous work on the analytical extraction of transport parameters. Leung *et al.* reported the transport properties of YAG:Ce³⁺ plates using a filtered broadband light source, where the linear dependence of ℓ_{tr} was exploited to calculate ℓ_a [54]. Firstly, the approximation used to analyze the total transmission was employed outside its range of validity. Secondly, their described method is only valid in the region of strong absorption or emission, and not in the overlap region. Hence the described approach could not be employed to extract transport properties over the whole visible spectral range.

Gaonkar *et al.* [91] used phenomenological theory (Kubelka-Munk theory) to decouple scattering and absorption properties of the diffusive samples. In the described approach the transport parameters are extracted using only an empirical relation between the Kubelka-Munk and the radiative transfer equation. Moreover, they extracted only the transport mean free path ℓ_{tr} and the absorption mean free path ℓ_a , which gives no explicit information about the anisotropy factor μ .

In Ch. 3 (published earlier as Ref. [86]) we reported the transport properties of YAG:Ce³⁺ plates using a narrowband light source that works in the whole visible spectral range including the overlap range ($\lambda_\ell = 490 \text{ nm} < \lambda_i < \lambda_r = 520 \text{ nm}$). We calculated ℓ_a under the assumption that ℓ_{tr} linearly depend on the wavelength in the absorption range, as was confirmed earlier on. Nevertheless, in the strongly absorbing region, the employed diffusion theory is not valid. Hence this method cannot adequately capture the parameter range of white LEDs (see Fig. 4.1).

4.7 Summary and outlook

We have investigated the transport of light through slabs that scatter and strongly absorb, with a focus on white LEDs. I extracted the transport parameters of white LED phosphor plates within the visible wavelength range. We measured total transmission, scattered reflection, and ballistic transmission over the whole visible wavelength range. I analyzed the diffuse light measurements using the P3 approximation to the radiative transfer equation to extract the scattering cross-section σ_s , the absorption cross-section σ_a and the anisotropy factor μ , in scattering media with strong absorption, up to $d/\ell_a = 4$. The Lambert-Beer law was used to analyze the ballistic light measurements, and extract $\sigma_s + \sigma_a$ in the whole visible spectral range. I verified the data obtained with the diffuse light measurements to the data measured with the ballistic light, and Monte Carlo simulations. Our study confirms the validity of the P3 approximation for light propagation in a medium suitable for white LEDs (see Fig. 4.1).

The procedure of finding transport parameters is a significant hindering step for the conventional light propagation modeling, such as ray tracing [15–18, 76]. Our method avoids the main bottleneck of these conventional numerical methods, providing at least 17-fold speed-up in time with the same error (see Fig. 4.8). The described approach is a suitable analytical tool for the industry since it provides a fast yet accurate determination of key transport parameters, and more importantly, it introduces predictive power into the design process of white LEDs. Both the know-how of and the control over optics that strongly scatters light has dramatically advanced [23–28, 92]. The state-of-the-art is much less developed regarding optical systems that also strongly absorb light (or even re-emit light of a different color), even though essential application fields occur in this regime. Examples are biomedical optics [32–37], or powder technology [38–40]. In this chapter, we provide a tool that also can be used in the fields listed above.

Chapter 5

Predicting the color of white LEDs

5.1 Introduction

In spite of the fast-growing market penetration of white LEDs devices, there are only a trial and error approaches for their design. Here we make a paradigm shift in white LED design, by introducing a radically new design principle for white LED. Our design principle predicts the color point [55] as a function of the design parameters and shows excellent agreement with our experimental observations. The proposed method allows exploring a large parameter space as it pertains to any phosphor and any polymer used in white LED production. Our model provides for the fast and efficient design, resulting in a reduction of both design and production costs. Widespread adoption of our design approach will lead to a dramatically increased proliferation of efficient white light sources, even in remote locations with little technological infrastructure.

The main characteristic of a white light source is the color point that is described using color coordinates in the color space shown in Fig. 5.1 [93,94]. The rim of the color space indicates pure monochromatic colors, and space is filled with all mixtures of monochromatic colors. By choosing the proper mix of monochromatic colors, one designs the color point of any white light source. Optical designers in an industry currently use extensive numerical simulations [95,96] to extract the color point from the design parameters of the white light source. Conversely, to target a specific color point, optical designers use a trial and error Monte Carlo ray tracing approach. To systematically design the color point of a white LED one needs to connect the design parameters of a white LED to the color point.

To adjust the color point one can control several design parameters such as the phosphor layer thickness d , the phosphor concentration ρ , the phosphor type, the type of blue LED, the additional scattering elements. In this chapter we demonstrate an analytical method to set the design parameters of a white LED starting from a targeted color point or conversely,

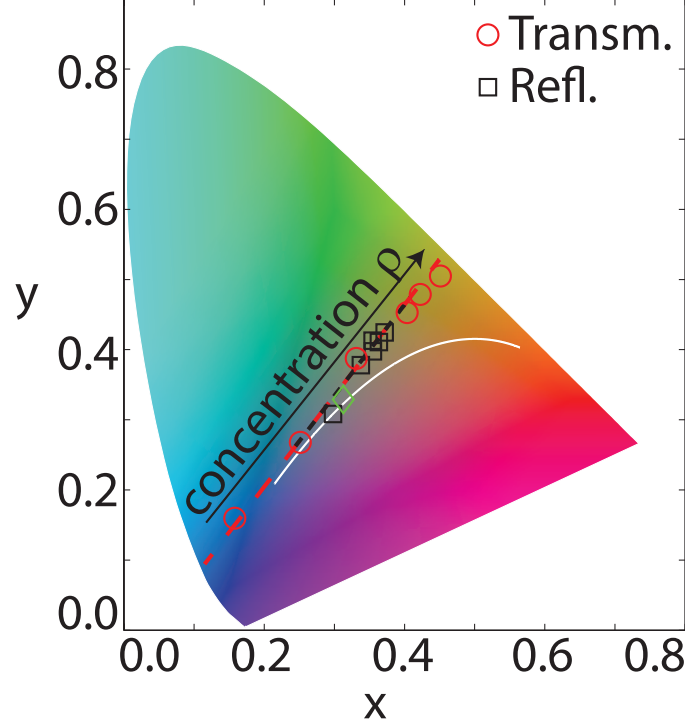


Figure 5.1: **Color point of a white LED.** The colored area indicates x-y color space [93]. Red and black dashed lines represent predicted color point dependence (curves in Fig. 5.4) on the phosphor concentration ρ (1 wt% to 8 wt%) for transmitted and reflected light, respectively. Circles and squares are our experimental data points. The green diamond indicates the most widely used standardized white light spectrum - D65 spectrum. The white curve represents the Planckian locus of blackbody temperature.

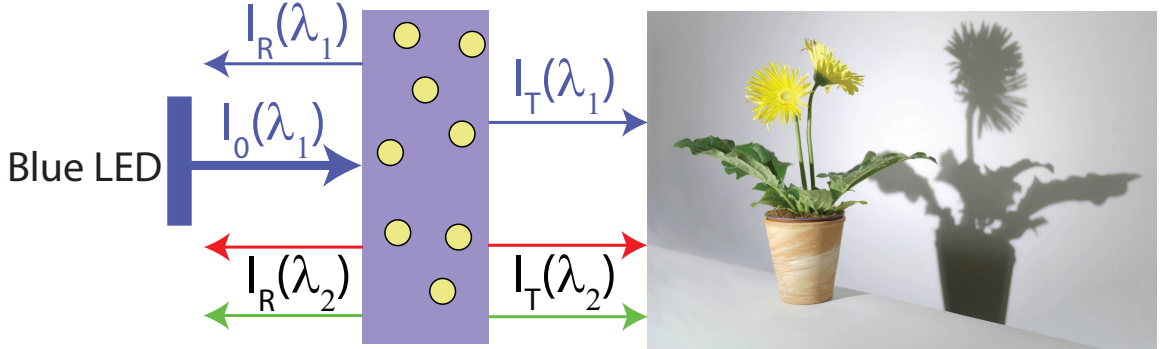


Figure 5.2: **Model for light propagation in a white LED slab.** Incident blue light with flux $F_0(\lambda_1)$ is shone by the blue LED on the phosphor slab with thickness d . The phosphor slab contains phosphor microparticles that are represented by yellow circles. $F_T(\lambda_1)$ is the total transmitted flux, $F_R(\lambda_1)$ is the scattered reflected flux, $F_T(\lambda_2)$ is the transmitted re-emitted flux, and $F_R(\lambda_2)$ is the reflected re-emitted flux. The mixture of transmitted red, green and blue light illuminates the object.

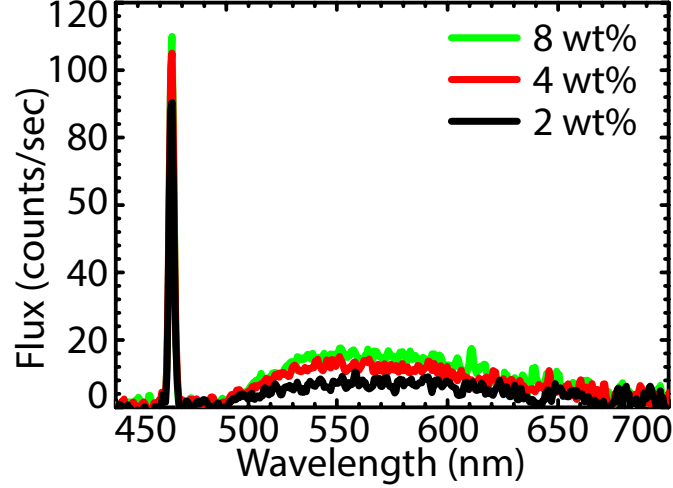


Figure 5.3: **Representative measured signal of a model white LED.** The sharp peak at $\lambda_1 = 475$ nm is the scattered incident light in reflection $F_R(\lambda_1)$, and the broad peak between 490 nm and 700 nm is light re-emitted by the phosphor $F_R(\lambda_2)$. Our model is not limited to the choice of the blue LED wavelength.

to predict the color point of a white LED starting from the chosen parameters. Based on the developed model we derived the expressions for the total flux emitted in the forward direction by model white LED with reflecting mirror

5.2 Methods

Our analytical model describes all light fluxes emanating from a white LED (see Fig. 5.2). We study light propagation through a model white LED that consist of a polymer diffuser plate containing YAG:Ce³⁺ phosphor microparticles. The refractive index of the diffuser plates is $n = 1.4$, and the plate thickness $d = 1.98 \pm 0.02$ mm. The absorption and the emission spectra of the phosphor are discussed in Ch. 3. The phosphor particle concentration ρ ranges from 1 wt% to 8 wt%. A tunable blue light source with flux $F_0(\lambda_1)$ illuminates the samples at a wavelength λ_1 see Ch. 3. The incident light is scattered, absorbed, and re-emitted resulting in the transmitted blue flux $F_T(\lambda_1)$, the reflected flux $F_R(\lambda_1)$, the re-emitted flux in transmission $F_T(\lambda_2)$, and the re-emitted flux in reflection $F_R(\lambda_2)$, where λ_2 is the wavelength of the re-emitted light. The transmitted and reflected fluxes are separately collected with an integrating sphere and detected with a fiber spectrometer.

The typical signal of our model white LED is shown in Fig. 5.3. The sharp peak at $\lambda_1 = 475$ nm originates from the blue source and contributes to the light fluxes $F_T(\lambda_1)$ and $F_R(\lambda_1)$. The broad peak between 490 nm and 700 nm consists of the light fluxes $F_T(\lambda_2)$ and $F_R(\lambda_2)$. From the experiment we derive the following quantities that will be analytically

modeled, namely the total transmission $T(\lambda_1) = F_T(\lambda_1)/F_0(\lambda_1)$, the re-emitted transmission $T_r(\lambda_1, \lambda_2) = F_T(\lambda_2)/F_0(\lambda_1)$, the scattered reflection $R(\lambda_1) = F_R(\lambda_1)/F_0(\lambda_1)$, and the re-emitted reflection $R_r(\lambda_1, \lambda_2) = F_R(\lambda_2)/F_0(\lambda_1)$. The transmissions $T(\lambda_1)$, $T_r(\lambda_1, \lambda_2)$ and the reflections $R(\lambda_1)$, $R_r(\lambda_1, \lambda_2)$ are obtained as a ratio of the areas under the scattered and re-emitted signal, as shown in Fig. 5.3.

5.3 Results and Discussion

The experimental results for the scattered blue light are shown in Fig. 5.4 (insets). The transmission $T(\lambda_1)$ of the scattered light reveals a sharp decrease with the phosphor concentration ρ as a result of scattering and strong absorption of the incident light. The reflection $R(\lambda_1)$ hardly varies with the phosphor concentration due to absorption; if the phosphor did not absorb light, we would have observed growth of the reflection with the increasing phosphor concentration, but the absorption counters this increase.

The measured transmission $T_r(\lambda_1, \lambda_2)$ and reflection $R_r(\lambda_1, \lambda_2)$ of the re-emitted light are shown in Fig. 5.4 (a)-(b). At low concentration, the re-emitted light flux $T_r(\lambda_1, \lambda_2)$ increases with the phosphor concentration, because more phosphor absorbs more light. Saturation occurs at $\rho = 3.3$ wt% because a further increase of the concentration increases the probability of a photon being absorbed very close to the entrance surface of the scattering material. When most photons are absorbed near the entrance surface, then most re-emitted photons are back reflected resulting in a decrease of the transmitted re-emitted flux, and an increase of the reflected re-emitted flux.

The transmission $T(\lambda_1)$ and reflection $R(\lambda_1)$ of the scattered light are analytically calculated using the P3 approximation [33, 35, 36] to the radiative transfer equation (see Ch. 2) [42]. Input parameters of the solution are the slab thickness d , the particle concentration ρ , the scattering cross section $\sigma_s(\lambda_1)$, the absorption cross section $\sigma_a(\lambda_1)$ and the anisotropy factor $\mu(\lambda_1)$ of the phosphor particles. The optical designer chooses the first two parameters, and the last three parameters can be obtained for the whole visible spectral range with an experimental method described elsewhere [91, 97] or from prior knowledge on the phosphors [41]. To describe the re-emitted light we calculate the absorbed average light flux distribution $U(z)$ inside the slab [43] (see Fig. 5.4 (c)). The average flux $U(z)$ is used as a source function for the re-emitted light that is also modeled by the P3 approximation [98] weighted by the quantum yield $q(\lambda_1, \lambda_2)$. The quantum yield is the ratio of the

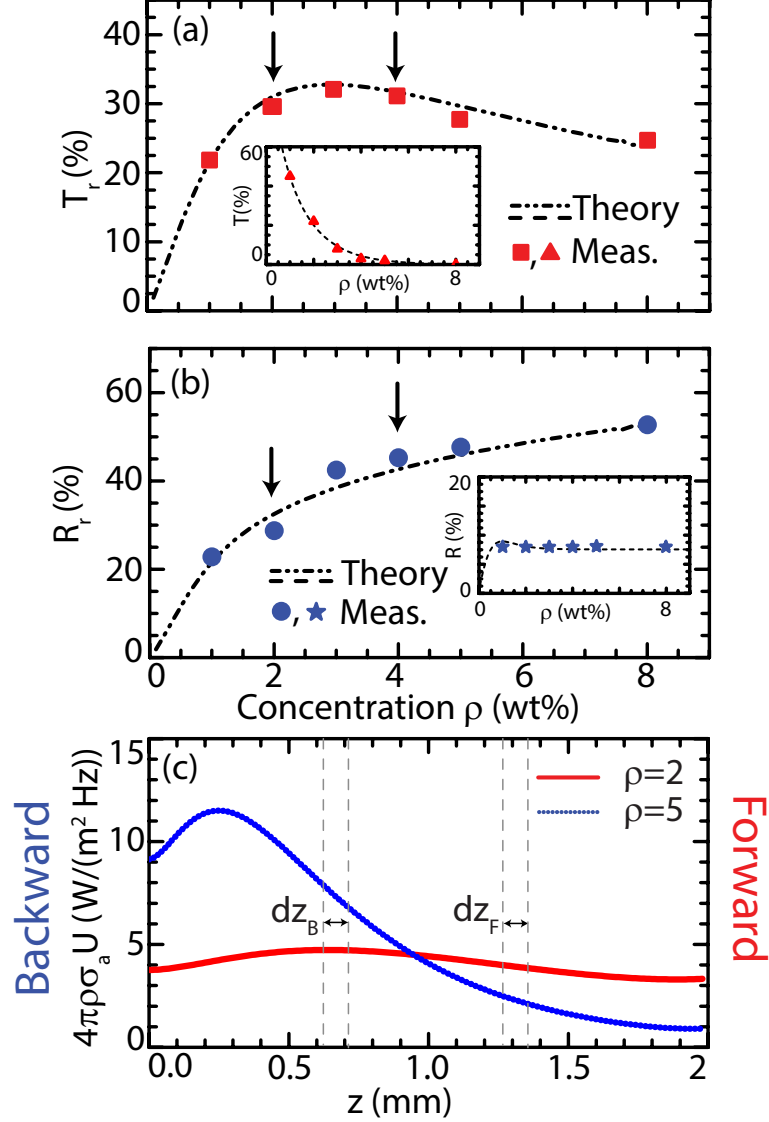


Figure 5.4: **Transmission and reflection as a function of phosphor particle concentration.** (a) Squares represent the measured transmission of the re-emitted light, dashed line represent predicted transmission of the re-emitted light. (b) Circles represent the measured reflection of the re-emitted light, dashed-dot-dot line represent predicted reflection of the re-emitted light. The error bars of the experiment are within the symbol size, and here $T_r = \int_{490 \text{ nm}}^{700 \text{ nm}} F_T(\lambda) d\lambda / F_0(\lambda_1)$, $R_r = \int_{490 \text{ nm}}^{700 \text{ nm}} F_R(\lambda) d\lambda / F_0(\lambda_1)$. (c) Lines represent the average flux $U(z)$ as a function of depth inside the sample with thickness 2 mm for two different phosphor particle concentrations ρ indicated with the black arrows in panels (a)-(b). Two representative ranges highlighted with gray dashed lines are shown at $d/3$ and $2d/3$ depth in the sample. Colored labels "Backward" and "Forward" indicate the backward and forward light propagation directions. The insets in panels (a)-(b) show data and theory for the scattered light, compare Fig. 4.7.

total number of re-emitted photons to the total number of absorbed photons [99, 100]

$$q(\lambda_1, \lambda_2) \equiv (T_r(\lambda_1, \lambda_2) + R_r(\lambda_1, \lambda_2)) / (1 - (T(\lambda_1) + R(\lambda_1))). \quad (5.1)$$

We extracted quantum yield $q(\lambda_1, \lambda_2)$ using data shown in Fig. 5.4 (a-b). The broadband measurement of the quantum yield $q(\lambda_1, \lambda_2)$ is shown in (Fig. 5.5). We see that quantum yield increases when we shift to the higher wavelength. The quantum yield saturates at a wavelength $\lambda = 444$ nm, and oscillates around the value of ~ 0.85 , comparable to the previously reported values [101, 102].

For the re-emitted light the P3 approximation to the radiative transfer equation is solved to extract the transmission $T_r(\lambda_1, \lambda_2)$ and reflection $R_r(\lambda_1, \lambda_2)$. Input parameters include the same parameters for the scattered light mentioned above, in addition the scattering cross section $\sigma_s(\lambda_2)$, the absorption cross section $\sigma_a(\lambda_2)$, the anisotropy factor $\mu(\lambda_2)$, and the quantum yield $q(\lambda_1, \lambda_2)$ of the phosphor particles.

The results of analytical solutions of the P3 approximation for the transmissions $T(\lambda_1)$ and $T_r(\lambda_1, \lambda_2)$ and the reflections $R(\lambda_1)$ and $R_r(\lambda_1, \lambda_2)$ as function of concentration ρ are shown in Fig. 5.4 (a)-(b). We find an excellent agreement between the experiment and our analytic model. To explain the behaviour of the $T_r(\lambda_1, \lambda_2)$ and $R_r(\lambda_1, \lambda_2)$ as a function of phosphor concentration we plot the absorbed power $4\pi\rho\sigma_a U(z)$ as a function of depth inside the scattering medium in Fig. 5.4(c), where the z coordinate corresponds to the notations in Fig. 5.2. When the concentration of the phosphor particles is low, the absorbed power is distributed almost uniformly across the sample. Re-emitted light generated as a result of absorption in a representative layer dz_B contributes mostly to the re-emitted reflection $R_r(\lambda_1, \lambda_2)$ because the light that is re-emitted in the backward direction in this layer will experience less scattering compared to the forward re-emitted light. The re-emitted light in a second representative layer dz_F will mostly contribute to $T_r(\lambda_1, \lambda_2)$ for the same reason. When the phosphor concentration is high, the flux is mostly absorbed near the entrance surface of the slab. Hence, layer dz_B will contribute a higher flux in the backward direction compared to the low concentration case. The layer dz_F will contribute less flux in transmission $T_r(\lambda_1, \lambda_2)$ compared to the low concentration case. This interplay results in the peak in transmission $T_r(\lambda_1, \lambda_2)$ at $\rho = 3.3$ wt% and a steady increase of reflection $R_r(\lambda_1, \lambda_2)$ with concentration ρ .

The color points for the measured data and our model are plotted in the color space in Fig. 5.1. The theoretical curves indicate all possible color points that can be achieved with the given YAG:Ce³⁺ phosphor when the concentration ρ is changed. The observed dependence of the color point on concentration is remarkably linear given the observation above Fig. 5.4 that the scattered and re-emitted light fluxes depend non-linearly on the concentration. The color point linearly shifts to the yellow part of the color space with increasing concentration for both the transmitted and reflected light in Fig. 5.1. The reason

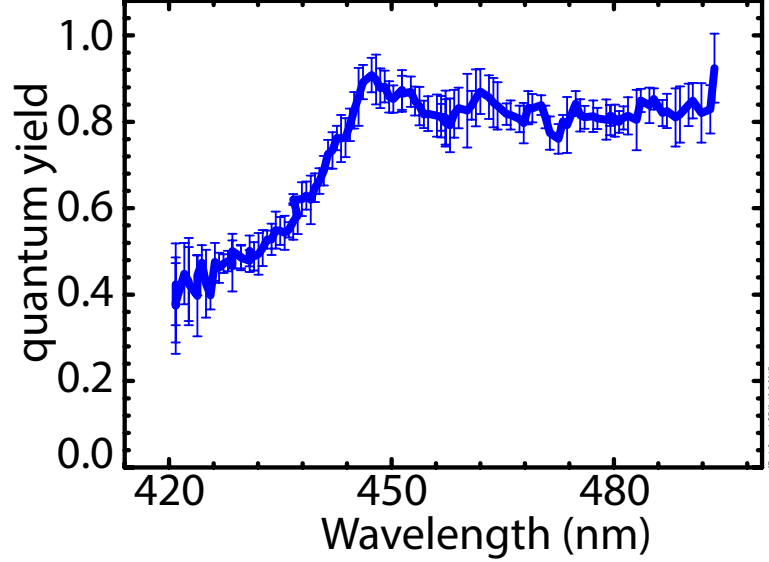


Figure 5.5: **Quantum yield of YAG:Ce³⁺**. Broadband quantum yield as a function of wavelength extracted from our measurements.

is that a higher concentration enhances the absorption of blue light increasing the produced re-emitted light.

5.4 White LED with backreflector

As a next step, we present a more complex model of a white LED that is relevant for several practical devices. In the commercially available white LED semiconductor and polymer are mounted in the cup covered with a thin layer of silver. Silver works as a mirror that reflects backscattered light back into the system and enhances the efficiency of the light source.

To account for this silver layer in our model, we place the mirror with reflectivity R_m on the left side of the polymer slab (see Fig. 5.6(b)). This mirror reflects the backscattered light at all wavelength. To calculate the resulting light flux emitted by the white LED we derive equations for the light flux using the iterative procedure. In the first stage, we illuminate the polymer slab with the excitation light flux F_0 . The light is being scattered, absorbed and re-emitted by the phosphor in all directions. In Fig. 5.6(b) dashed box of stage 1 scattering encloses all resulting fluxes produced by the incident flux F_0 . The excitation light flux emitted in the backward direction is equal to $F_0 R$, where R is the reflectance of the polymer slab. The re-emitted light flux emitted in the backward direction is equal to $F_0 R_r$, where R_r is the reflectance of the polymer slab for the re-emitted light. The fluxes exciting on the right side of the sample are $F_0 T$, and $F_0 T_r$ for excitation and re-emission respectively, where T is the transmission of the slab and T_r is the transmission of the re-emitted flux.

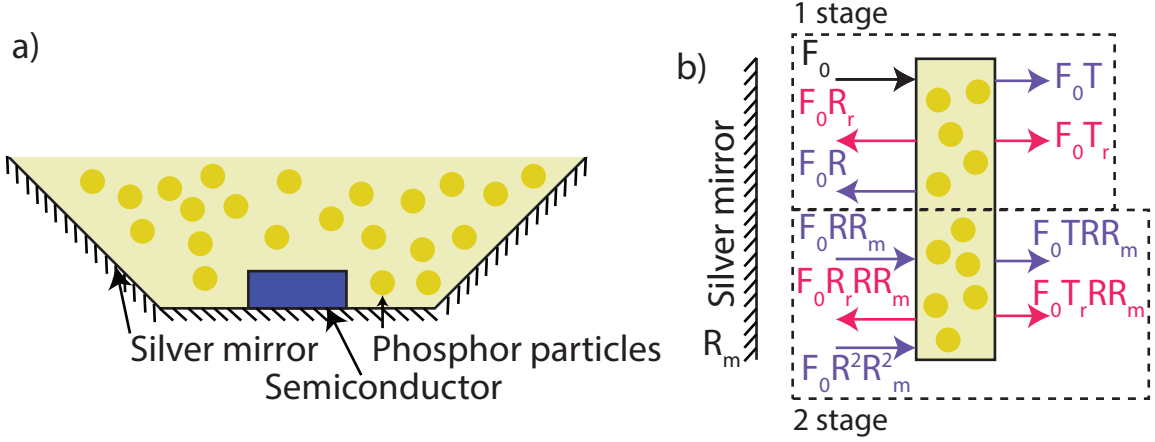


Figure 5.6: **White LED with back-reflector.** (a) Scheme of the white LED with back-reflector. Blue rectangle is a semiconductor. Yellow circles are the phosphor inclusions. On the bottom and the sides of the LED cup silver mirror is located. (b) Silver mirror reflects the excitation light and the re-emission light that was backscattered by polymer plate with phosphor particles. R is the reflectivity of the excitation light, R_r is the reflectivity of the re-emitted light. Red arrows represent the re-emitted flux, and blue arrows represent the excitation flux. Dashed boxes enclose stage 1 and 2 of the scattering and re-emitting processes.

The part of the backscattered light flux $F_0 R R_m$ is being reflected from the mirror and again illuminates the slab. This is the second stage of the scattering that is indicated in Fig. 5.6(b). The transmitted fluxes are $F_0 T R R_m$ and $F_0 T_r R R_m$ for excitation and re-emission, respectively. The backscattered fluxes are $F_0 R^2 R_m^2$ and $F_0 R_r^2 R_m^2$ for excitation and re-emission, respectively. This process of reflection from the mirror, scattering and re-emission continue to infinity. For the excitation light, the total transmitted light flux is calculated as

$$F_{\text{tot}} = F_0 T + F_0 T R R_m + F_0 T R^2 R_m^2 + \dots = F_0 T \sum_{n=0}^{\infty} R^n R_m^n = F_0 T \frac{1}{1 - R R_m}, \quad (5.2)$$

where we used the equation of the geometric sum. In the limit of the perfect mirror ($R_m = 1$), and non-absorbing particles ($\sigma_a = 0$) Eq. 5.2 becomes

$$F_{\text{tot}} = F_0 \frac{T}{1 - R} = F_0, \quad (5.3)$$

where we used the fact that in the non-absorbing medium $T = 1 - R$. All the light eventually gets through the sample.

The transmitted re-emitted light is calculated in the same way as transmitted excitation

light

$$F_T^{\text{re-em}} = F_0 T_r + F_0 T_r R R_m + F_0 T_r R^2 R_m^2 + \dots = F_0 T_r \sum_{n=0}^{\infty} R^n R_m^n = F_0 T_r \frac{1}{1 - R R_m}, \quad (5.4)$$

we replaced T with T_r in this equation. This is not all re-emitted flux that is emitted on the right side of the sample. The backreflected re-emitted flux F_b is being reflected from the mirror and partially transmitted without absorption on the right side of the sample. To calculate the total flux emitted on the right side of the sample we need to account for this contribution. In order to calculate this contribution we first calculate the total backreflected re-emitted flux F_b . The total backreflected re-emitted light flux is calculated in the same way as Eq. 5.4 by replacing T_r with R_r

$$F_b = F_0 R_r + F_0 R_r R R_m + F_0 R_r R^2 R_m^2 + \dots = F_0 R_r \sum_{n=0}^{\infty} R^n R_m^n = F_0 R_r \frac{1}{1 - R R_m}, \quad (5.5)$$

where R_r defines the amount of the re-emitted light generated inside the slab and emitted in the backward direction. The generated re-emitted light flux F_b is being reflected by the mirror ($F_b R_m$), and propagates back and forth through the sample with no absorption $\sigma_a = 0$. The transmitted part of flux F_b^{tr} can be calculated using Eq. 5.4 by replacing F_0 with $F_b R_m$

$$F_b^{\text{tr}} = F_b R_m T_R + F_b R_m T_R R R_m + F_b R_m T_R R^2 R_m^2 + \dots = F_b T \sum_{n=0}^{\infty} R^n R_m^n = \frac{F_b R_m T_R}{1 - R R_m}, \quad (5.6)$$

where T_R and R_R are the transmission and reflection of the light at the wavelength of the re-emitted light. Finally we sum all the contributions Eq. 5.4 and Eq. 5.6 for transmitted re-emitted flux

$$F_{\text{tot}}^{\text{re-em}} = F_b^{\text{tr}} + F_T^{\text{re-em}} = \frac{F_0 T_R R_r R_m}{(1 - R_R R_m)(1 - R R_m)} + \frac{F_0 T_r}{1 - R R_m}, \quad (5.7)$$

where $F^{\text{re-em}}$ is the re-emitted light generated on the right side of the slab due to every passage of the excitation light through the slab and $F_{\text{tot}}^{\text{re-em}}$ is the total flux of the re-emitted light emitted on the right side of the slab. When we remove mirror from the system ($R_m = 0$) Eq. 5.7 results in

$$F_{\text{tot}}^{\text{re-em}} = F_T^{\text{re-em}} = F_0 T_r, \quad (5.8)$$

that is equal to the first passage of F_0 through the slab. In another extreme when the

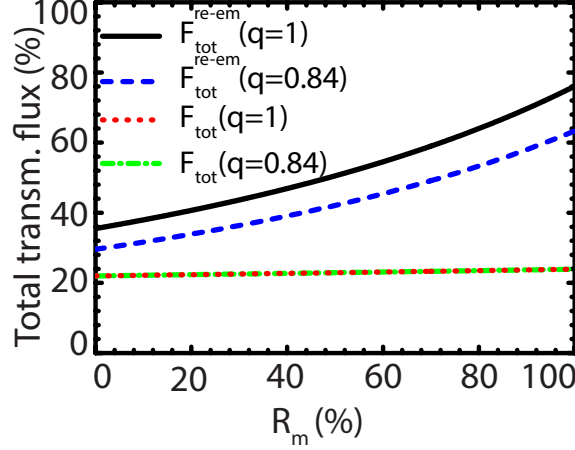


Figure 5.7: **Total transmitted flux as a function of mirror reflectivity.** Transmitted flux of the scattered and re-emitted light is shown for two values of quantum yield. Total transmitted flux for the re-emitted light depends on the quantum yield. The transmitted flux for the scattered light does not depend on the quantum yield of the phosphor.

mirror is perfect $R_m = 1$ and the quantum yield $q = 1$ ($A = T_r + R_r = 1 - T - R$, where A is the absorbed light) all incident flux is transmitted in the forward direction

$$\begin{aligned}
 F_{\text{tot}} + F_{\text{tot}}^{\text{re-em}} &= \frac{F_0 T}{1 - R} + \frac{F_0 \cancel{T_R} R_r}{(1 - \cancel{R_R})(1 - R)} + \frac{F_0 T_r}{1 - R} = \frac{F_0 T}{1 - R} + \frac{F_0 (R_r + T_r)}{1 - R} = \\
 &= \frac{F_0 T}{1 - R} + \frac{F_0 (1 - T - R)}{1 - R} = F_0, \quad (5.9)
 \end{aligned}$$

where we used the equality $T_R + R_R = 1$. This equality follows from the assumption that the re-emitted light is not absorbed by the phosphor as justified in Ch. 4.

In Fig. 5.7 we plotted Eq. 5.2 and Eq. 5.7 as a function of the mirror reflectivity R_m for two values of the quantum yield q . We see that up to $\sim 34\%$ point of additional re-emitted flux is redirected in the forward direction when a mirror is used in the system $R_m = 1$ compared to no mirror $R_m = 0$. The re-emitted flux depends considerably on the quantum yield q and mirror reflectivity R_m of the system, while on the contrary, scattered flux does not depend on the quantum yield q and insignificantly depend on the mirror reflectivity R_m . This disproportional increase of the re-emitted and scattered flux results in the shift of the color point to the red part of the color triangle. The derived expressions for fluxes can be used for any excitation and emission wavelength λ , outside the overlap region described in Ch. 3.

5.5 Summary

We have developed an analytical model that can revolutionize white LED design. It provides a simple, efficient design tool, hence, allowing to access a large design parameter space using an analytical algorithm without adjustable parameters. While our current solution offers a one-dimensional description of the light propagation, the analytical solution of the P3 approximation in 3D geometry [45] is already available and can describe light propagation in complex geometries that are typically employed in white LED design. Additionally, we obtained the expression for the spectrum of a white LED including the back-reflector. Here we derived the expressions for the total flux emitted in the forward direction by model white LED with a reflecting mirror. Our model showed that the use of the mirror provides an increase of the total transmitted flux up to $\sim 34\%$ point and shifts the color point of the LED to the red part of the spectrum. Our approach will increase the design efficiency by avoiding recurring design efforts and decrease the cost of ownership of white LEDs units for worldwide users.

Chapter 6

Universal limit on the applicability of diffusion theory for light propagation

6.1 Introduction

Diffusion theory is one of the most widely used theories for describing physical processes of heat diffusion, electron transport, sound waves and light propagation. It is an approximation to more general transport theory. Diffusion theory for light fail when applied to those special systems in which the scattering is predominantly in the forward direction, or in which absorption is extreme. The precise range of applicability of diffusion theory has never been established. Here we present a fundamental, universal limit beyond which diffusion theory of light gives unphysical results when applied to the scattering materials. In the unphysical range, the errors in transport parameters can reach thousands of percents.

6.2 Diffusion theory

Understanding the physics of light in scattering materials with absorption and different degree of anisotropy is crucial in many areas of research, ranging from astrophysics to biology and sustainable energy generation [28, 32–40, 80–84, 103, 104]. The transport theory is used to describe how light propagates in these complex materials [43]. Given the complexity of transport theory, an extremely popular approximation to transport theory is the diffusion theory of light [43].

Diffusion theory has been successfully applied to describe experimental observations on numerous materials with widely varying optical properties such as clouds [105], biological tissue [106], quantum dots [107], powders [108], and white LEDs [97]. Diffusion is supposed

to be a valid approximation to transport theory for a broad range of transport parameters that characterize scattering media [105, 106, 109]. The accepted ranges of validity are typically described by comparing a sample thickness d to several characteristic length scales [77]:

$$\ell_{\text{tr}} \ll d \ll \ell_{\text{a}}, \quad (6.1)$$

where the transport mean free path ℓ_{tr} is the average distance light travels before losing information about its initial direction due to scattering, and the absorption mean free path ℓ_{a} is the average distance light travels before being absorbed by $1/e$ [23, 28, 63, 92].

Whereas the inequalities (6.1) provide guidelines for the range of validity, they do only offer a qualitative limit. It has been shown that in some instances the diffusion equation still yields reasonable predictions, although the inequalities (6.1) were violated [110]. Hence, the central questions of our paper are: Is there a sharply defined range of validity of diffusion theory? What happens outside such a possible range? In a nutshell, the answer provided by this chapter are: yes, there is a sharp range of validity, and outside these ranges, we found unphysical behavior.

Both transport theory and diffusion theory describe light using the characteristic scattering length scales $(\ell_{\text{s}}, \ell_{\text{a}}, \ell_{\text{tr}})$. The scattering mean free path ℓ_{s} is the average distance light travels between two consecutive collisions. These characteristic length scales have straightforward relations to the single-particle properties $(\sigma_{\text{s}}, \sigma_{\text{a}}, \mu)$, where σ_{s} is the scattering cross section of the particles constituting the scattering material, σ_{a} where is the absorption scattering cross section of the particles, and μ , is defined as the average scattering angle $\langle \cos \theta \rangle$ of a scattering particle. These relations are [43]

$$\ell_{\text{s}} = \frac{1}{\rho \sigma_{\text{s}}} \equiv \frac{1}{\mu_{\text{s}}}, \quad (6.2)$$

$$\ell_{\text{a}} = \frac{1}{\rho \sigma_{\text{a}}} \equiv \frac{1}{\mu_{\text{a}}}, \quad (6.3)$$

$$\frac{1}{\ell_{\text{tr}}} = \frac{(1 - \mu)}{\ell_{\text{s}}} + \frac{1}{\ell_{\text{a}}} \equiv \frac{1}{\mu'_{\text{s}}} + \frac{1}{\mu_{\text{a}}}, \quad (6.4)$$

where ρ is the density of scattering particles. The introduced coefficients μ_{s} , μ_{a} , μ'_{s} are often used in biological tissues studies [106, 111, 112]. The Eqs. (6.2-6.4) are essentially the result of expansions of scattering theory in inverse density $1/\rho$, and their validity goes far beyond transport theory [43].

In widely-used geometries, such as a finite slab or a semi-infinite slab, the diffusion

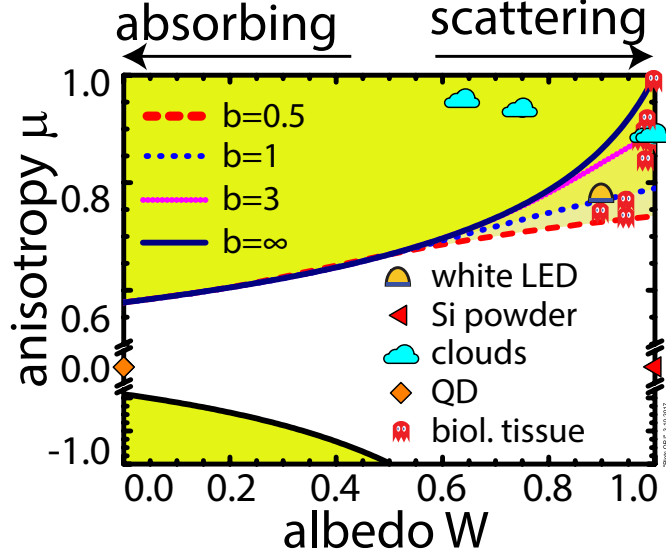


Figure 6.1: **Validity range of diffusion theory for a slab shown as a 2D map.** Lime-green regions are the areas where the diffusion equation gives unphysical results. The dashed and dotted curves show the boundary of validity for different values of b . The solid lines indicate the boundary for a semi-infinite slab ($b = \infty$). The data symbols represent sample parameters for popular scattering media, as described in the legend [66, 97, 105–107].

equation allows for simple, analytical solutions. For instance, for a plane wave incident upon a finite slab with thickness d the diffusion equation for the diffuse energy density $U_d(z)$ has the form [43]

$$\frac{\partial^2}{\partial z^2} U_d(z) - \kappa_d^2 U_d(z) = -Q_0 \exp(-\rho \sigma_t z), \quad (6.5)$$

where $Q_0 = [3\rho\sigma_s\rho\sigma_{tr} + 3\rho\sigma_s\rho\sigma_t\mu](F_0/4\pi)$, F_0 is the magnitude of the incident flux, $\sigma_{tr} = (1 - \mu)\sigma_s + \sigma_a$ is the transport cross section, and $\kappa_d^2 = 3\sigma_a\sigma_{tr}$. The mixed boundary conditions are

$$U_d(z) - h \frac{\partial}{\partial z} U_d(z) + \frac{Q_1(z)}{2\pi} = 0 \quad \text{at } z = 0, \quad (6.6)$$

$$U_d(z) + h \frac{\partial}{\partial z} U_d(z) - \frac{Q_1(z)}{2\pi} = 0 \quad \text{at } z = d, \quad (6.7)$$

where $Q_1(z) = \sigma_s\mu\sigma_{tr}F_0 \exp(-\rho\sigma_t z)$, $\sigma_t \equiv \sigma_s + \sigma_a$ is the total cross section, and $h \equiv 2/(3\rho\sigma_{tr})$. The solution to differential equation Eq. (6.5) with boundary conditions (6.6-6.7) is textbook material [43]. As this solution is the starting point of our discussion, we have verified this solution both analytically and numerically. The diffuse energy density

$U_d(z)$ is a scalar, positive definite quantity rigorously expressed as

$$U_d(z) \geq 0 \quad \forall z \in [0, d]. \quad (6.8)$$

The flux $\mathbf{F}(z) \equiv F(z)\hat{\mathbf{z}}$ is a one-dimensional vector [43], whose direction, changes from leftward at the front surface to rightwards at the back surface of the slab, expressed rigorously as

$$F(z) < 0 \quad \text{at } z = 0, \quad (6.9)$$

$$F(z) > 0 \quad \text{at } z = d, \quad (6.10)$$

the total diffuse flux directed inwards must be zero. The rigorous conditions (6.8-6.10) replace the inequalities (6.1). When any of these conditions (6.8-6.10) is violated the diffusion equation fails, as experimentally observable quantities would take on unphysical values, such as negative reflection or transmission coefficients, or a negative energy density.

To check the validity of the diffusion equation for the slab and the semi-infinite slab we explored the full parameter space. Several independent sets can fully characterize the sample. We have chosen the set consisting of the three dimensionless parameters (W, μ, b) , where the albedo W is defined as $W \equiv \sigma_s/(\sigma_s + \sigma_a)$, and where the dimensionless optical thickness of the slab b is defined as $b \equiv d/\ell_s$.

6.3 Results

Our main results are summarized in Fig. 6.1. The two lime-green regions indicate that part of the parameter space where diffusion theory fails, as it generates negative energy density $U_d(z)$. We discovered that the conditions on the flux $F(z)$ and the energy density $U(z)$ are equivalent. In Fig. 6.1 the two yellow-green boundaries of the applicability range of the diffusion theory were calculated from the following expressions:

$$U_d(0) = 0, \quad \text{at the right side of the slab,} \quad (6.11)$$

$$U_d(d) = 0, \quad \text{at the left side of the slab.} \quad (6.12)$$

The equivalent conditions for the fluxes are formulated as follows:

$$F(0) = 0, \quad \text{at the right side of the slab,} \quad (6.13)$$

$$F(d) = 0, \quad \text{at the left side of the slab.} \quad (6.14)$$

Naively, one would expect the boundaries defining the validity of the diffusion equation to depend on the optical density. However, we find that only the top region depends significantly on the optical thickness, as shown in Fig. 6.1. When the albedo $W > 0.5$, the colored area of the top region is more significant for the greater optical thickness b . When $W < 0.5$, we observe no changes in the top colored area for different optical thicknesses b . This insensitivity for strongly absorbing samples can be explained by the fact that light never reaches the backward boundary due to the strong absorption.

We explore some limiting cases relevant for experiments. In case of systems without absorption ($W = 1$) Eqs. (6.11-6.12) simplified to the following condition on the anisotropy parameter μ for the diffusion equation to have physical solutions

$$\mu \leq \frac{-1 + 3b + \exp(-b)}{3b}. \quad (6.15)$$

In the additional limit of vanishing optical thickness we obtain

$$\mu \leq \frac{2}{3} \quad \text{at } b \rightarrow 0, W = 1, \quad (6.16)$$

for the diffusion approximation to hold. The limit $b \rightarrow \infty$ and $W = 1$ gives physical results for all values of μ . A slab with optical thickness $b > 10$ can for all practical purposes be considered to behave as a semi-infinite slab.

From an experimental point of view, a scattering sample is typically characterized with the set of length scales $(d, \ell_{\text{tr}}, L_a)$, where the absorption length L_a is the average distance between the start and end of a random-walk with absorption mean free path ℓ_a . We emphasize that the set $(d, \ell_{\text{tr}}, L_a)$ will always be a valid set characterizing light transport through a sample, irrespective of the validity of diffusion theory.

6.4 Error map of the diffusion theory

It is an experimentally relevant question how scientists who study samples that lie in the unphysical according to Fig. 6.1 regions of diffusion theory yet still interpreting the results with diffusion theory without encountering any problem. How is this possible? In a typical experiment, the total transmission coefficient T and total reflection coefficient R are measured. From these experimental data, the lengths $(\ell_{\text{tr}}^{\text{d}}, L_a^{\text{d}})$ are inferred *using diffusion theory*, and no unphysical behavior is found for samples having (b, W, μ) far in the unphysical regions. In such a case the extracted values $(\ell_{\text{tr}}^{\text{d}}, L_a^{\text{d}})$ correspond to values for (b, W, μ) that differ strongly from the true values (b, W, μ) for those samples. We will now investigate

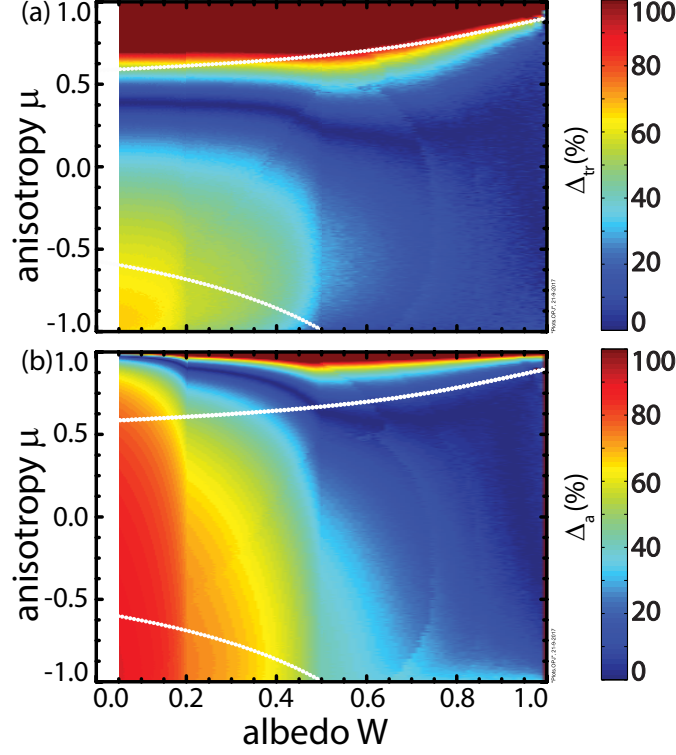


Figure 6.2: **Relative error in diffusion theory calculations shown as a 2D map.** The white curves show the analytically derived boundary of the diffusion theory validity range for the optical thickness $b = 3$. (a) Color gradient indicates the relative error Δ_{tr} in the obtained mean free path ℓ_{tr}^d using the diffusion theory compared to the rigorous ℓ_{tr} . (b) Color gradient indicates the relative error Δ_a in the obtained absorption length ℓ_a^d using the diffusion theory compared to the rigorous ℓ_a .

how much they differ.

To gauge the errors associated with the diffusion approximation we have performed extensive Monte Carlo simulations. The simulation procedure is described in details in Ref. [97]. For the simulations samples are characterized by the set (b, ℓ_{tr}, L_a) . For several values of b we calculate the transmission T and the reflection R for many values of the pair (W, μ) and convert this set onto the (ℓ_{tr}, L_a) space. So we have an exact mapping of (ℓ_{tr}, L_a) to (T, R) and vice versa for a specific value of b . Given the set (T, R) obtained from our simulations we can calculate *with diffusion theory* values for the length scales: (ℓ_{tr}^d, L_a^d) that would produce these values for T and R . These approximate values should be compared with the rigorous values (ℓ_{tr}, L_a) .

To evaluate the relative error of the length scales obtained with diffusion theory we define relative errors

$$\Delta_{tr} \equiv \frac{|\ell_{tr} - \ell_{tr}^d|}{\ell_{tr}^d}, \quad (6.17)$$

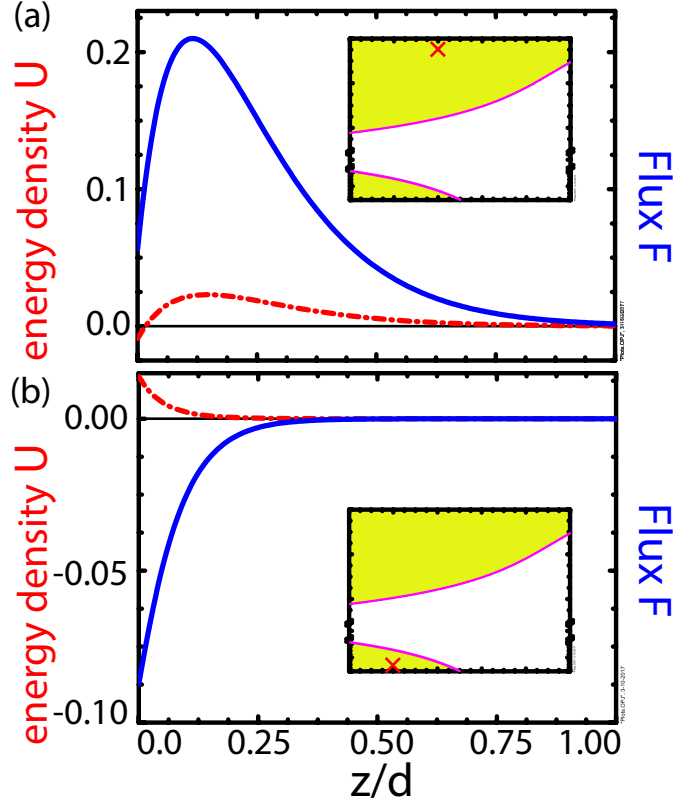


Figure 6.3: **Energy density (red curves) and flux (blue curves) in the unphysical range of transport parameters.** (a) Example of a strongly forward scattering and absorbing material where the unphysical conditions on the left boundary of the slab $F(0) > 0$ and $U(0) < 0$ hold. (b) Example of a strongly backward scattering material with strong absorption where the unphysical conditions at the right boundary of the slab $F(d) < 0$ and $U(d) < 0$ hold. Insets in both graphs are similar to Fig. 6.1. Red crosses indicate parameters of the samples in (a) and (b). Results are shown for $b = 3$. Energy density is scaled with $F_0/4\pi$, and the flux is scaled with F_0 , F_0 being the magnitude of the incident flux.

and

$$\Delta_a \equiv \frac{|L_a - L_a^d|}{L_a^d}. \quad (6.18)$$

In Fig. 6.2 we show the 2D map of the relative errors for $b = 3$ for both Δ_{tr} and Δ_a . First, we consider the range of albedo $W \geq 0.5$. For this albedo range the relative errors Δ_{tr} and Δ_a do not exceed 10% inside the major part of the physical area. In the narrow range close to the analytical boundary relative errors for the ℓ_{tr} can reach up to 80% for low albedo W in the physical region. The relative error for ℓ_a is less than 10% near the analytical boundary, but shows higher error up to 40% near the $\mu = -1$. For $W = 1$ we see a narrow red region for all values of μ in Fig. 6.2. Diffusion theory with absorption always erroneously predict (ℓ_{tr}^d, L_a^d) for $W = 1$. Inside the unphysical region, the relative error Δ_{tr}

exceeds 100% and Δ_a exceeds 10% for the albedo mentioned before.

Secondly we consider the albedo range of $W \leq 0.5$, where the inequality (6.1) is violated. In this region the diffusion theory results in relative errors larger than 50%, except for a narrow (blue) island that can be seen in the left part of Fig. 6.2(a) and an island in Fig. 6.2(b). The magnitude of errors in these two regions does not exceed 20%. These two islands do not overlap, meaning that if we identify one parameter from the pair (ℓ_{tr}^d, L_a^d) correctly, the corresponding another parameter will have errors exceeding 50%. Without any prior knowledge about the sample, we cannot guess what quantity from the pair (ℓ_{tr}^d, L_a^d) is extracted correctly. To conclude, diffusion theory gives unreliable results when interpreting data on samples with $W \leq 0.5$, because the error of transport parameters can reach thousands of percent.

In Fig. 6.2 we have indicated with white lines the analytical boundaries of the validity of the diffusion theory, for the case of $b = 3$. These boundaries divide the scattering parameter space into three parts. The diffusion theory results in physical solutions only in the middle region. For $W \geq 0.5$ the white line separates the area with the high error from the part with low error obtained with Monte Carlo simulations. For $W \leq 0.5$ diffusion theory results in significant errors as discussed before.

To indicate more in detail the wrong predictions of the diffusion theory from the invalid region, we show in Fig. 6.3 the energy density and flux for two samples with correct scattering parameters. Transport parameters of sample 1 and sample 2 are shown in the inset of Fig. 6.3(a)-(b) respectively. In Fig. 6.3(a) the flux and energy density show unphysical behavior near the left boundary of the sample. For the transport parameters in Fig. 6.3(b) the flux and energy density show unphysical behavior near the right boundary of the sample. The relative errors of the length scales are $(\Delta_{tr} = 1056\%, \Delta_a = 115\%)$ and $(\Delta_{tr} = 56\%, \Delta_a = 74\%)$ for sample 1 and 2 respectively.

6.5 Summary

In conclusion, we have derived the fundamental limit of diffusion theory in media with absorption and anisotropic scattering. We identified a set of three parameters consisting of the anisotropy factor, the albedo, and the optical thickness that together fully characterizes any multiply scattering material with or without absorption. It is pointed out that in the space defined by this parameter set there are large areas where the diffusion equation gives unphysical results, such as negative energy densities. We demonstrated that when diffusion theory is used to infer mean free paths and absorption lengths from experimental data,

relative errors in these length scales can be as large as thousands of %.

Chapter 7

Reversible tuning of the color point of a white LED

7.1 Introduction

White light is conveniently characterized by a color point that is represented on the color space shown in Fig. 7.1 (see Appendix). The color point of a white LED depends on its design parameters (see Ch. 5). For example in Fig. 7.1 we show how the color point depends on the phosphor particle concentration. When the design parameters are chosen, the color point of the white LED cannot be changed. Here, we show that wavefront shaping technique can be employed to *reversibly tune* the color point of a white LED without changing the design parameters (see Fig. 7.1).

7.2 Wavefront shaping

Wavefront shaping defines how the light travels through the scattering medium. It exploits the interference effects inside the scattering medium in such a way that the scattering medium can even be transformed into a lens [22, 113, 114]. The plane wave that is incident on the scattering medium is diffused while undergoing multiple scattering inside the slab in the absence of wavefront shaping. However, when we change the phase or the amplitude of the incoming wavefront with the help of wavefront shaping, the light can be focused into the single focus spot behind the sample (see Fig. 7.2). To achieve the focusing effect, the incident wavefront is optimized using algorithms described in detail in Ref. [115]. The quality of the wavefront is gauged using the enhancement η , which is defined as

$$\eta \equiv \frac{F_{\text{opt}}(\lambda)}{\langle F_{\text{rand}}(\lambda) \rangle}, \quad (7.1)$$

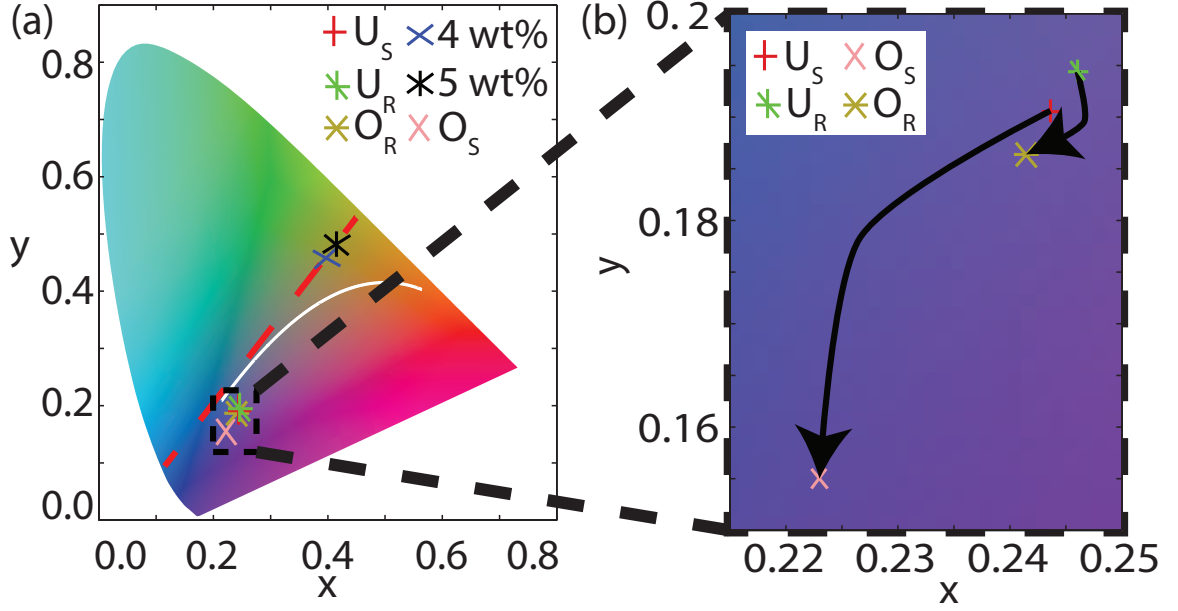


Figure 7.1: **Tuning of the color point of a white LED.** (a) Color triangle with the color points obtained under different conditions. The dashed red line represents the color point as a function of concentration for the given phosphor. Red crosses correspond to the color point of the scattered light before and after wavefront optimization. Green crosses correspond to the color point of the re-emitted light before and after optimization. Black and dark blue crosses correspond to the color point of the diffuser plates with different phosphor concentrations [97]. (b) The dashed inset shows enlarged area with the color points extracted from the experiment.

where $F_{\text{opt}}(\lambda)$ is the flux of the diffused light with an optimized wavefront and $\langle F_{\text{rand}}(\lambda) \rangle$ is the flux of the diffused light with an unoptimized or random wavefront.

Here we exploit wavefront shaping to change the color point of a white LED reversibly. There are two main differences between the wavefront shaping approach described above and one that is used in this work. The first difference is that the wavefront shaping is done here on a large area of the order of mm. We measure all outgoing light from the sample. The second difference is that the phosphor particles that are used in white LEDs not only scatter light but also absorb and re-emit part of it in the red part of the spectrum. Hence, the optimization can be done for the scattered light and re-emitted light. Here we optimize either on the scattered light or the re-emitted light.

Having these two differences in mind, we define two enhancements, one for the scattered light (η_s) and one for the re-emitted light (η_r)

$$\eta_{s,r} \equiv \frac{\int_{\lambda_1}^{\lambda_2} F_{\text{opt}}(\lambda) d\lambda}{\int_{\lambda_1}^{\lambda_2} \langle F_{\text{rand}}(\lambda) \rangle d\lambda}, \quad (7.2)$$

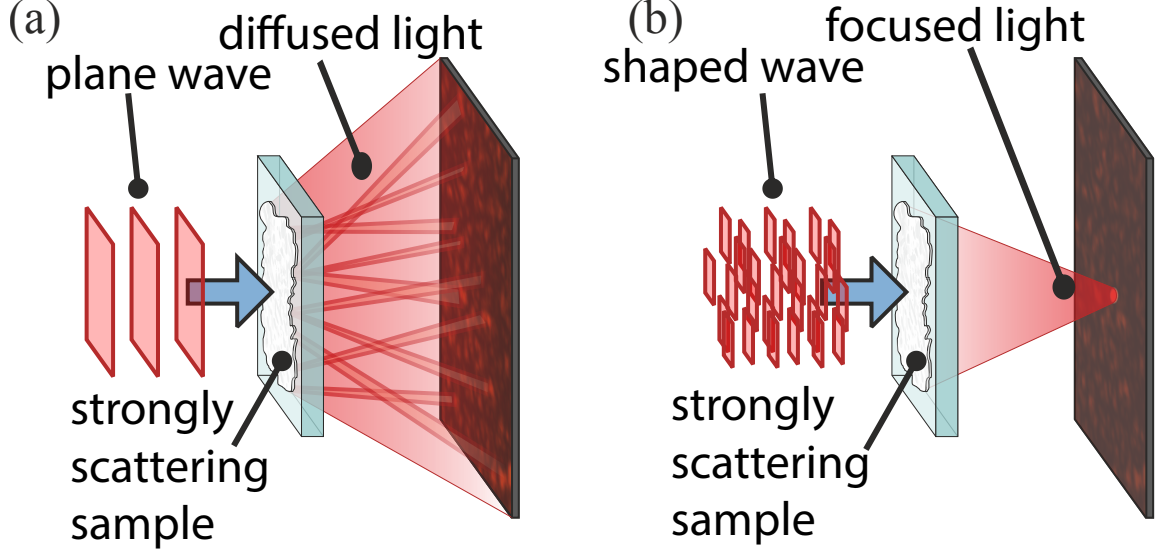


Figure 7.2: **Wavefront shaping technique.** (a) A plane wave is incident on a scattering medium. The light is multiply scattered resulting in the transmitted diffuse light. (b) By applying a certain wavefront, the light is scattered in the medium in such a way that it focuses behind the scattering medium. Figure adopted from [116].

where $F_{\text{opt}}(\lambda)$ is the flux at wavelength λ integrated over the area of the diffused spot when an optimized wavefront is used, $F_{\text{rand}}(\lambda)$ is the flux at wavelength λ integrated over the area of the diffused spot when an unoptimized wavefront is used, and $\langle \dots \rangle$ denotes the average over many random wavefronts. The integration is performed for the following range of wavelengths: for the scattered light we use $(\lambda_1 = 455 \text{ nm}, \lambda_2 = 460 \text{ nm})$ and for the re-emitted light we use $(\lambda_1 = 475 \text{ nm}, \lambda_2 = 700 \text{ nm})$ (see Fig 7.8).

The expected enhancement can be theoretically estimated for a multiple channel optimization in the following way [117]:

$$\eta = \frac{\pi}{4} \frac{N - 1}{M} + 1, \quad (7.3)$$

where N is the number of segments of the spatial light modulator (SLM), M is the number of modes in the system that depends notably on the wavelength, and the bandwidth of the light source. The number of modes M_s for the scattered light can be estimated to be $M_s = 2\pi A \arcsin(\text{NA})/\lambda^2$, where A is the area of the diffused spot, NA is the numerical aperture of the setup, and λ is the illumination wavelength [117]. The number of modes M_r for the re-emitted light is estimated as $M_r = 8\pi n_{\text{eff}}^3 V \Delta\lambda/\lambda^4$, where V is the volume of the diffused light cone inside the sample [118], $\Delta\lambda = \lambda_2 - \lambda_1 = 225 \text{ nm}$ is the full width at half maximum of the emission spectrum, $\lambda = 587.5 \text{ nm}$ is the central wavelength, and n_{eff}

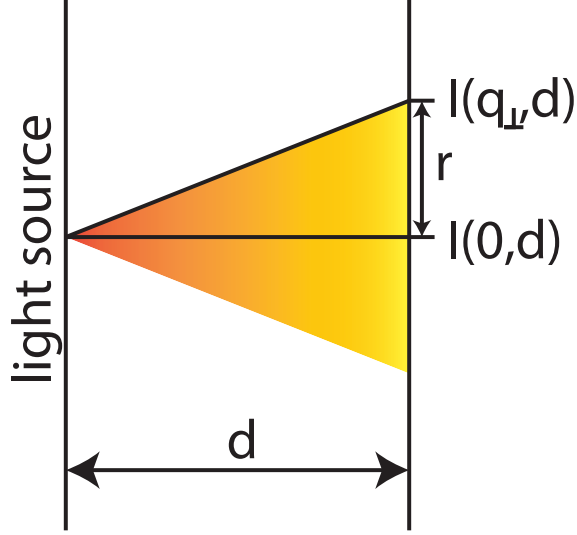


Figure 7.3: **Schematic of the diffused light cone inside a slab of scattering material.** The focused light creates point source on the left surface of the plate. Light diffuses inside the scattering medium, and forms the diffuse light spot with radius r on the right surface of the slab.

is the effective refractive index.

To calculate the number of modes M we need to estimate the size of the diffused spot created on the sample by the incident beam. To estimate the size of the diffused spot we use the expression for the diffuse flux using the 2D diffusion equation in Fourier k -space [117]

$$F_d(\mathbf{k}_\perp, z) = A \frac{\sinh(k_\perp[d_e - z - z_e]) \sinh(k_\perp[\ell_{tr} + z_e])}{k_\perp \sinh(k_\perp d_e)}, \quad (7.4)$$

where A is a constant that depends on the incident flux and on the diffusion constant, $d_e \equiv d + 2z_e$, $k_\perp \equiv \sqrt{k_{\perp x}^2 + k_{\perp y}^2}$ and $z_e = J_1 \ell_{tr} / J_0$, the constants J_0 and J_1 are defined in Ch. 2 Eq. 2.91. We define the radius of the diffused spot to be the solution of the following equation (see Fig. 7.3)

$$\frac{F_d(k_\perp, d)}{F_d(0, d)} = e^{-2}. \quad (7.5)$$

Since Eq. 7.5 is a transcendent equation, it cannot be solved exactly. For the diffuser plate used in the experiment the numerical solution of Eq. 7.5 yields $k_\perp = r = 1.28$ mm. From here we calculate the area of the diffused spot $A = \pi r^2 = 5$ mm², and the number of modes of the scattered light to be $M_s = 10^8$. The mode volume of the re-emitted light is calculated to be $V = \pi r^2 d / 3 = 3.43$ mm³, so the number of modes for the re-emitted light is huge: $M_r = 4.3 \times 10^{11}$. Typically in wavefront shaping experiments, the number of modes is of the order of 10^3 for the scattered light, which allows gaining substantial control over

light in the experiment.

7.3 Setup

In this chapter, we discuss the setup based on a setup that was constructed to demonstrate wavefront shaping to a broad audience robustly. Hence parameters of the setup were optimized for demonstration purposes. The experimental setup is shown in Fig. 7.4. The light source is a blue diode laser (Roithener Lasertechnik, rl450-40-5) with a wavelength $\lambda = 457$ nm to excite the phosphor. The beam is expanded to fill the active area of the digital micromirror device (DMD). The original beam shape of the laser is elliptical with dimensions 3 by 5 mm². The beam is expanded by 6.7 times in the horizontal direction using two plano-convex cylindrical lenses (C1 and C2) to illuminate a higher number of segments on the DMD. Mirror M1 and M2 direct the beam onto a DMD. The DMD (Vialux V-7001, DLP700) has the resolution of 1024x786 pixels and a pixel size of 13.68 μm . The angle of incidence of light on the DMD should be set to 24 degrees to control the pixels of the DMD. The on-position of the pixel corresponds to the micromirror position that reflects light in the direction of lens L1, and off-position of the pixel corresponds to the location of the micromirror that reflects light to the beam dump (CB). We apply the superpixel method [119] using lenses L1, L2 and the aperture (A). The lenses both have a focal length 150 mm and are placed 300 mm apart in a 4f configuration. In the focal plane of L1, the aperture (A) blocks the first diffraction order. Mirrors M3 and M4 are used to steer the beam. The beam is focused by a 75 mm lens (L3) onto the sample (S). The sample is silicone (PDMS) plate with phosphor particles (YAG:Ce³⁺) with a concentration of YAG:Ce³⁺ particles of 4 wt % made by Jan Jansen at Philips Lighting. The sample has a circular shape with the diameter of 6 cm and is 2 mm thick. The optical characterization of the sample is described in detail in Chapter 4.

The sample is placed in the focal plane of the lens L3. A 2-inch lens L4 with focal length 35 mm is positioned at a distance of 39 mm from the sample. The beam is split using a 92/8 pellicle beam splitter. The feedback arm receives 8% of the light and 9% of the light is sent to the spectrometer arm. In the feedback arm, the light is spectrally filtered using a short-pass (FES0500) or long-pass (FEL0500-1V1-45-11) filter F to optimize on the scattered or the re-emitted light respectively. The cutoff wavelength of the filter is $\lambda = 500$ nm. An image of the diffused spot on the sample is projected on the charged coupled device camera (CCD, Allied Vision Guppy PRO F-146) placed at distance 320 mm from L4 (01LAGO13). A feedback system FB processes the images taken by the CCD and sent it to the DMD. In

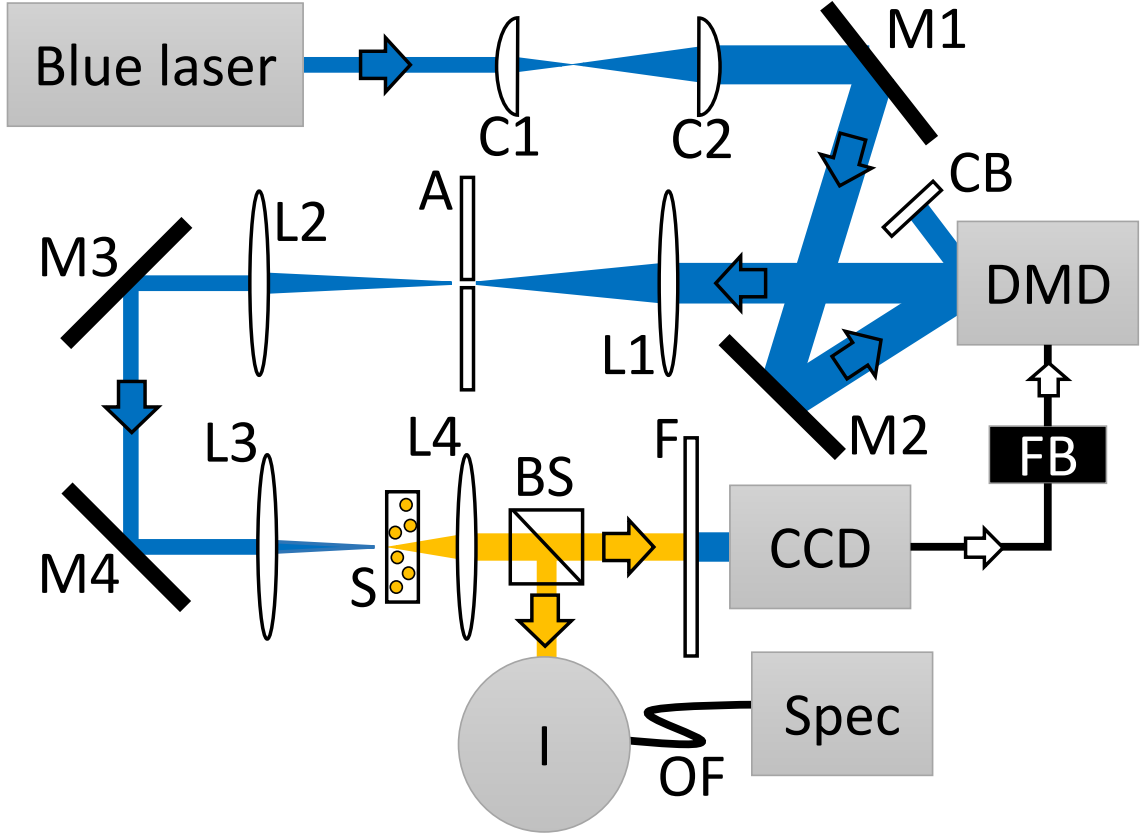


Figure 7.4: **A schematic of the setup for wavefront shaping on a phosphor sample.** The blue laser beam is expanded and modulated by the digital mirror device (DMD). With a series of lenses and mirrors, the beam is focused onto the sample surface. The flux is transmitted through the sample and collected by the lens L4, and split by the beam splitter between CCD camera and integrating sphere.

the spectrometer arm, an integrating sphere I is placed at the same distance (320 mm) to L4 as the CCD. In this way, we ensure that the collected flux is the same for the CCD and the integrating sphere. The spectrum of light is detected by a spectrometer (Spec, Avantes AvaSpec-ULS2048XL-EVO) connected to the integrating sphere through the optical fiber OF. The setup allows to perform wavefront shaping optimization and acquire the spectrum of emitted light simultaneously.

The feedback system is controlled by a LabView program that optimizes a wavefront using the partitioning algorithm [115]. The optimization procedure starts by applying a random wavefront to the DMD and taking an image of the speckle pattern. Then 50 % of the segments are randomly selected and shifted 3 times with the phase shift $\pi/4$. Generated wavefront is sent back to the DMD, and a new image is acquired with the CCD. This process is repeated over and over again until the desired wavefront is obtained. The optimization

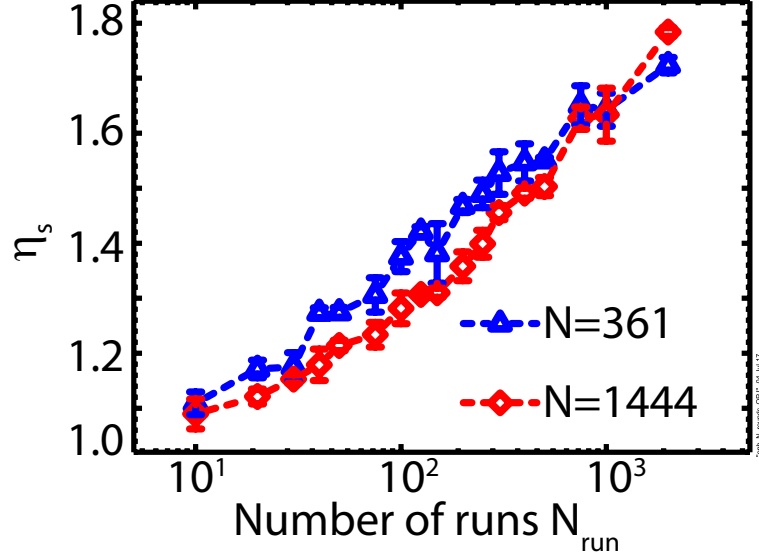


Figure 7.5: **The wavefront enhancement registered on the CCD as a function of the number of optimization rounds N_{run} .** Each data point is averaged over 3 measurement rounds; the error bars are the standard deviations. The number of segments used during the optimization is indicated in the legend.

process takes approximately 30 min to achieve enhancement of 2.

7.4 Characterization of the setup

7.4.1 Optimization algorithm

For optimal performance of the optimization algorithm, we need to identify the number of segments N used on the DMD, and the number of optimization runs N_{run} . Here we find experimentally the number of runs required for optimization. The enhancement η on the CCD is measured as a function of N_{run} for two different values of total controlled segments ($N = 361$ or 1444). The segments are arranged in a 19 by 19 or a 38 by 38 square respectively. The illuminated area of the DMD is independent of the number of pixels used in the superpixel.

The measured results are shown in Fig. 7.5. We see that at a large number of runs N_{run} an increase in the number of controlled segments results in a higher enhancement. When N_{run} is smaller than 10^3 the enhancement is higher for a lower number of segments. When N_{run} is greater than 10^3 the enhancement is higher for a higher number of segments. Hence we conclude that for the higher number of the segments it takes longer to optimize a wavefront, but the optimized flux is higher than for optimization with the small number of segments. For the measurements performed in this report the total number of segments N is

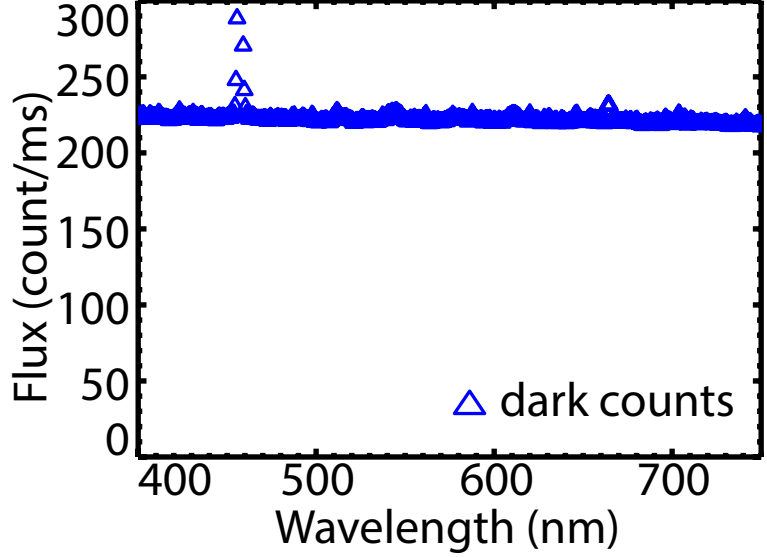


Figure 7.6: **Dark counts measurement for the experimental setup.** The standard deviation of the data points is within the symbol size. The prominent peak at $\lambda = 457$ nm is arising from stray light in the setup.

chosen to be 900 (30 by 30 segments) and the number of runs is selected to be $N_{\text{run}} = 2000$. We use segments of the sizes of 4 by 4 superpixels.

7.4.2 Detection system

In this section, we discuss the characterization of the detection system notably the dark counts and the sensitivity. To correct for the dark counts of the spectrometer, we measure the signal with an integration time $T_{\text{int}} = 500$ ms when the incident beam is blocked. The spectrum is averaged over 198 measurements and shown in Fig. 7.6. A peak at $\lambda = 457$ nm is observed, which coincides perfectly with the wavelength of the laser. The peak is caused by the stray light of the laser in the setup. Since this stray light is always present during the measurements, we take it into account. In this way, the stray light intensity is always subtracted from the real signal.

To calibrate the wavelength dependent sensitivity of the detection system, we measure a white LED that was characterized by Philips Lighting. Philips Lighting kindly provided a look-up table with light flux in watts for each wavelength emitted by white LED. The white LED light source is placed at the position of the sample, and the spectrum of the LED is recorded (see Fig. 7.7(a)). Standard spectrum F_c measured by Philips Lighting is compared to the signal F_m measured with our setup, so we can derive the calibration curve

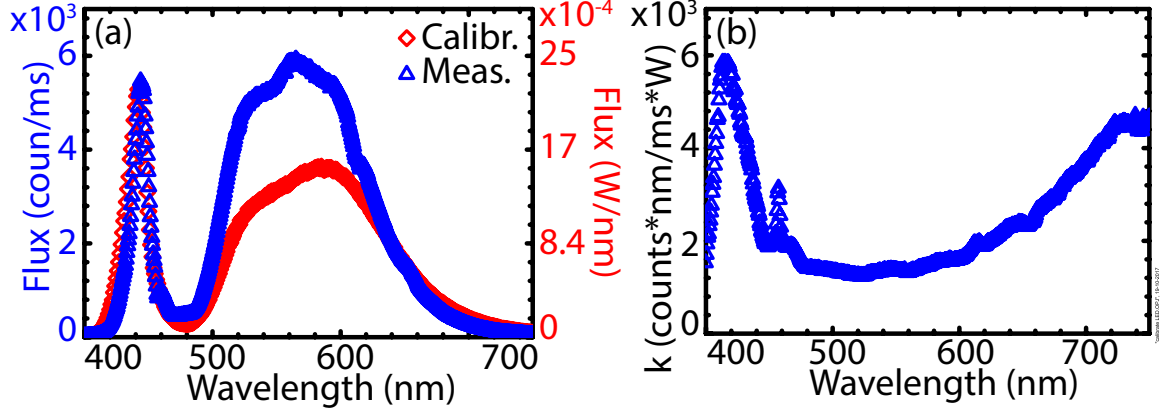


Figure 7.7: **Calibration of the measurement setup.** (a) The red spectrum is measured using calibrated equipment at Philips Lighting. The blue spectrum is measured in our setup. The ratio of these two spectra provides a calibration curve for our setup. (b) Calibration curve that is used to account for the wavelength dependence of the measurement setup.

$k(\lambda)$ of our setup using

$$k(\lambda) \equiv \frac{F_m(\lambda)}{F_c(\lambda)}, \quad (7.6)$$

The calibration curve is plotted in Fig. 7.7(b). We see a highly nonlinear normalization curve with peaks between 400 and 500 nm and a steady increase of $k(\lambda)$ with the wavelength that starts around 500 nm. The shape of this curve depends on the transmission characteristics of each component in the setup.

7.4.3 Measurement procedure

The blue laser illuminates the phosphor sample. The transmitted light that contains scattered and re-emitted light is registered by the spectrometer and CCD as described in Sec. 7.3. The CCD registers the total flux emitted by the sample and cannot discriminate signal at multiple wavelengths. To discriminate the scattered and the re-emitted light on the CCD during the optimization run, we use long pass filter or short pass filter. The measurement procedure consists of the following steps: first, we place a long pass or short pass filter in front of the CCD to filter the scattered or the re-emitted part of the spectrum from the measured signal. The choice of the filter depends on the light signal that we want to optimize. To optimize on the scattered light we use a short pass filter with the cutoff wavelength of $\lambda = 500$ nm, and to optimize on the re-emitted light we take a long pass filter with the cutoff wavelength of $\lambda = 500$ nm. When the choice of the filter is made, we optimize the measured flux on CCD. The input parameters of the program are set to $N_{\text{run}} = 2000$, $N = 1444$ and the segment size is 4 by 4 superpixels. We record the spectrum

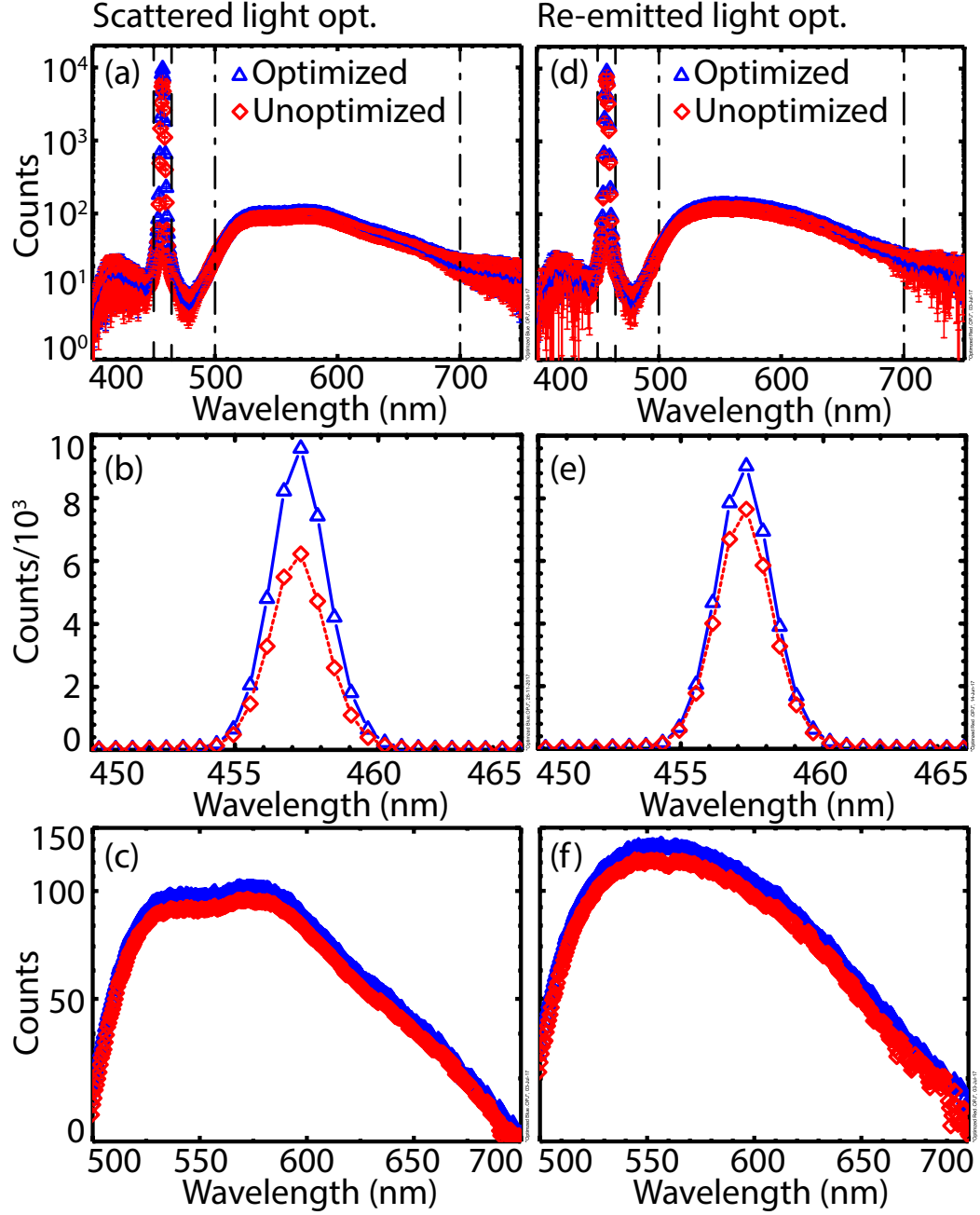


Figure 7.8: **Optimized spectra of the model white LED.** (a) Unoptimized and optimized spectrum measured when the optimization is performed on the scattered light. Dashed lines indicate the scattered light range, and dash-dotted lines indicate the re-emitted light. (b) Zoomed-in wavelength range of Fig. 7.8(a) of the scattered light. (c) Zoomed-in range of Fig. 7.8(a) of the re-emitted light. (d) Unoptimized and optimized spectrum measured when the optimization is performed on the re-emitted light. Dashed lines indicate the scattered light range, and dash-dotted lines indicate the re-emitted light. (e) Zoomed-in wavelength range of Fig. 7.8(d) of the scattered light. (f) Zoomed in range of Fig. 7.8(d) of the re-emitted light.

of the optimized wavefront and random wavefront shown in Fig. 7.9(a-b) and discussed in the next section. The integration time of the spectrometer is set to $T = 500$ ms. To calculate the standard deviation, we average over 99 measured spectra. The experiment is repeated 10 times for the scattered and for the re-emitted light to ensure reproducibility of the measured enhancement values. The achieved enhancement values are discussed in the next section.

7.5 Results and discussion

We illuminate the sample with the light source at the wavelength $\lambda = 457$ nm. The incident light is being scattered, absorbed and re-emitted inside the sample. The transmitted, scattered and re-emitted light is measured using an integrating sphere and CCD. Typical measured signals are shown in Fig. 7.8(a) and (d). The scattered transmitted light flux has a pronounced peak at $\lambda = 457$ nm that coincides with the wavelength of the light source, as expected. The re-emitted light flux is spread between $\lambda = 490$ nm and $\lambda = 700$ nm and coincides with the re-emission band of the phosphor.

We use a wavefront shaping algorithm to optimize the light flux of the scattered light. The spectrum that is optimized on the scattered light spectrum and plotted in Fig. 7.8(a). We see that the unoptimized spectrum has a lower flux at every wavelength in the visible range. To highlight the differences of the increased flux for the scattered and the re-emitted signal, we show the zoomed-in ranges of the spectrum in Fig. 7.8(b-c). In Fig. 7.8(b), we see the enhancement of the scattered light signal in the optimized case compared to the unoptimized case. The enhancement of the scattered light after optimization is $\eta_s = 1.48 \pm 0.03$. The similar enhancement of the re-emitted light is apparent in Fig. 7.8(c). The enhancement of the re-emitted light is $\eta_r = 1.074 \pm 0.007$. The enhancement of the re-emitted light is smaller than the enhancement of the scattered light. This difference in enhancements causes a shift of the color point of the transmitted light. We calculated the color point of the transmitted random and optimized light (see Fig. 7.1). The color point of the random wavefront is $x = 0.244, y = 0.191$, and for the optimized wavefront it is $x = 0.223, y = 0.155$. The error in the x and y coordinates is in the order of 1%. To quantify the color shift caused by wavefront shaping, we define the relative color shift Δ_d

$$\Delta_d \equiv \sqrt{\Delta x^2 + \Delta y^2}. \quad (7.7)$$

The color shift in our experiment is equal to $\Delta_d = 0.042 \pm 0.002$. For comparison, we

calculate the color shift that occurs when the mass concentration of the phosphor particles is changed from 4 wt % to 5 wt %. This color shift $\Delta_d = 0.03$ is taken as a reference, and we assign it to be 100%. Wavefront shaped color shift advances the concentration color shift by $136 \pm 7\%$. Hence, wavefront shaping technique allows to dynamically tune the color point without changing the phosphor concentration.

Next, we optimize the re-emitted light flux. The spectrum of the random and optimized light intensities is shown in Fig. 7.8(d). In Fig. 7.8(e-f) the enlarged parts of the spectra are shown for the scattered and the re-emitted light. Here we also observe an increase in the scattered and the re-emitted flux. The enhancement of the scattered light is $\eta_s = 1.26 \pm 0.09$, and the enhancement of the re-emitted light is $\eta_r = 1.10 \pm 0.02$. Although the optimization is done on the re-emitted light, the enhancement of the scattered light is higher than the enhancement of the re-emitted light. A possible reason might be that during the experiment more re-emitted flux is lost compared to the scattered flux. The re-emitted light has a uniform angular distribution, whereas the scattered light is more directional [97]. Hence, the collection objective with $\text{NA} = 0.65$ will collect predominantly forward scattered light and will miss light scattered at higher angles. Another possible reason is that the lower signal to noise ratio of the re-emitted light prevents the optimization process. During the experiment, we observed higher fluctuations of the re-emitted light flux compare to the scattered light flux. These fluctuations cause the optimization algorithm to perform worse [116]. We observed a difference in the shape of the re-emitted spectrum for the re-emitted light optimization and scattered light optimization (see Fig. 7.8(c) and (f)), however, the shapes of the spectra between optimized and unoptimized runs appear to be the same. The reason for such differences is currently unknown.

We calculated the color points of the unoptimized wavefront as $x = 0.246, y = 0.194$ and optimized wavefronts as $x = 0.241, y = 0.186$. The color point shift between optimized wavefronts and unoptimized wavefronts is $\Delta_d = 0.001 \pm 0.0005$, this corresponds to $3 \pm 2\%$ of the shift compared to the reference shift defined above. In Table 7.1 we summarize the obtained results. It is apparent from Table 7.1 that the color point shift for the re-emitted light is one order of magnitude smaller than for the scattered light.

We plot the enhancements η_s and η_r as a function of the diffused spot area for the scattered and re-emitted light (see Fig. 7.9). We used Ref. [117, 118] for the theoretical predictions of the number of modes. The theoretical curves decay very rapidly with the area of the diffused beam for both absorbed and the re-emitted light. For the re-emitted light even when we consider a typical μm^2 area, the theoretical enhancement is almost equal to 1 for our experimental setup and sample. Strikingly the experimental enhancement is

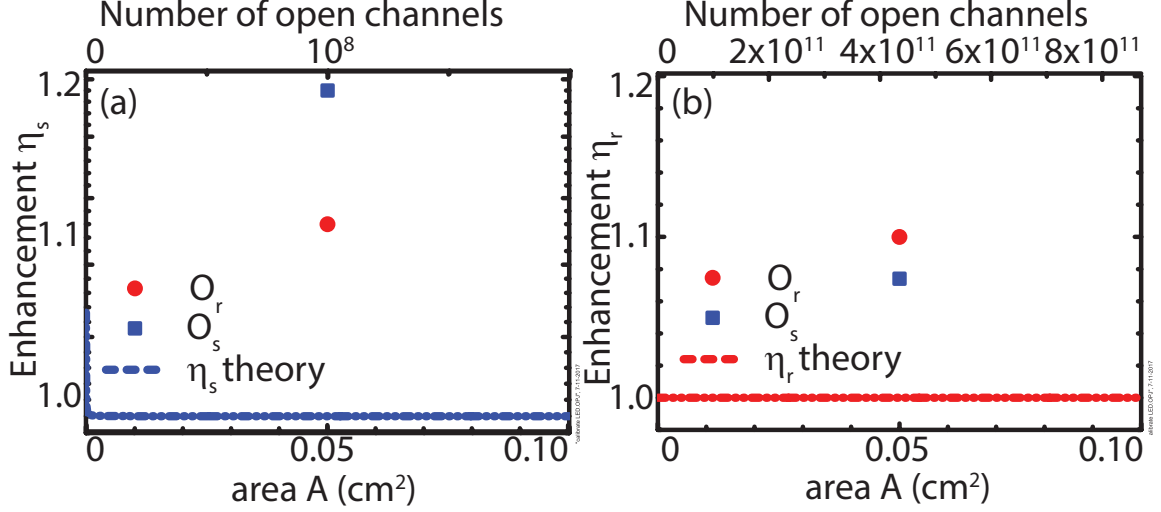


Figure 7.9: **Enhancement as a function of target area for scattered and re-emitted light.** (a) Enhancement as a function of the target area for the scattered light, where O_s is the enhancement of the scattered light and O_r is the enhancement of the re-emitted light. The data point is the measured value in this chapter. The top x axis indicates the number of open channels in the system. (b) Enhancement as a function of the target area for the re-emitted light. Theoretical curves were generated following the predictions of Ref. [117,118]

well above the theoretically predicted values. It implies that even with the few control channels and with a macroscopically large number of open channels, the transmitted flux can be increased. Because the increase of flux for the scattered and re-emitted light is not equal, we observe the color point shift of the emitted light.

Table 7.1: Overview of measurement results

	Optimized on scattered	Optimized on re-emitted
η_s	1.48 ± 0.03	1.26 ± 0.09
η_r	1.074 ± 0.007	1.10 ± 0.02
Δ_d	0.042 ± 0.002	0.001 ± 0.0005
% shift	136 ± 7 %	3 ± 2 %

7.6 Summary and outlook

We have demonstrated the tuning of the color point of a white LED using wavefront shaping technique. When optimizing on the scattered light we obtain enhancements $\eta_s = 1.48 \pm 0.03$ and $\eta_r = 1.074 \pm 0.007$. This results in a color point shift of $\Delta_d = 0.042$ that corresponds

to 36 % of the color shift due to irreversible concentration change. For optimization on the re-emitted light we find $\eta_s = 1.26 \pm 0.09$ and $\eta_r = 1.10 \pm 0.02$, resulting in a color point shift of $\Delta_d = 0.001$, which corresponds to 3% of the shift due to concentration change. We observed that the color point always shifts to the blue part of the spectrum which is caused by the fact that the enhancement of the scattered light is always bigger than the enhancement of the re-emitted light.

We demonstrated that the wavefront shaping could be achieved even on macroscopic samples ($A \approx 5 \text{ mm}^2$). In this chapter, we showed for the first time that enhancement on macroscopic objects in the re-emitted light could be achieved. The existing theories cannot describe the obtained enhancements. Ojambati *et al.* reported enhancements $\eta = 2$ on areas of $A = 15 \text{ }\mu\text{m}^2$, and it is surprising to see that an increase in the optimization area by 3 orders of magnitude resulted for our strongly absorbing samples in a comparable enhancement [115]. The enhancement of the transmitted flux demonstrated in this chapter paves the way to a white LED efficiency enhancement based on passive optical elements, such as the phase masks.

In conclusion in this chapter, we demonstrated the proof of principle reversible color tuning of a white LED. We have been able to wavefront shape light in a macroscopic area on strongly absorbing scattering samples with re-emission. We observed that both scattered and re-emitted light is enhanced in the process of wavefront shaping. The reached enhancements are comparable to the enhancements achieved in non-absorbing samples for μm^2 area.

Chapter 8

Summary

Lighting is a crucial technology that is used every day. The introduction of the white light emitting diode (LED) that consists of the Nobel Prize-winning technology of a blue LED combined with a phosphor layer, dramatically reduces the energy consumption for lighting. Despite the fast-growing market penetration, there are currently only trial and error approaches for white LED modeling. In this thesis, I define experimental and theoretical tools that analytically connect phosphor powder properties to the color point of a white LED. Phosphor particles inside the diffuser layer scatter, absorb and re-emit incident light. Absorption and scattering comparably affect light; hence, neither of the effects can be neglected. Due to these properties, widely used analytical tools fail to describe phosphor diffusion layers. I propose and experimentally verify a novel systematic approach that fully describes light diffusion and re-emission in phosphor layers.

The P3 approximation was chosen to be the central theoretical framework that allowed us to describe light propagation in scattering materials with strong absorption. This approximation is derived from the more general radiative transfer equation (RTE), and detailed steps of derivations are described in Chapter 2. The equations of the P3 approximation in the parallel slab geometry are analytically solvable, and presented at the end of the Chapter 2.

The P1 approximation is the first order approximation to RTE, and it has a much simpler analytical solution than the P3 approximation. In Chapter 6, it was shown that the P1 approximation gives a substantial error of the order of 30% in estimating transport parameters for the scattering materials with strong absorption, and it cannot be applied to some scattering materials. To identify the range of validity for the P1 approximation, I derive analytical expressions and 2D maps that provide error estimates of the P1 approximation for the whole transport parameter range.

To analyze data with the P1 and the P3 approximations, I performed measurements

in the range where scattered and re-emitted light are spectrally separated. Conventional measurement techniques fail to provide such separation when scattering materials with broadband re-emission are studied. In Chapters 3 and 4 I present a novel experimental setup that solves the problem. This setup uses narrowband illumination, so the scattered and re-emitted light is always spectrally separated or can be distinguished.

The central objective of Chapters 3 and 4 was to identify the transport parameters of the phosphor plates that are typically used for white LEDs. In Chapter 3, the extracted parameters were $(\ell_{\text{tr}}, \ell_{\text{a}})$, and the P1 approximation was used to extract these parameters. In Chapter 4, a more sophisticated experiment was performed, and it allowed to identify a wider parameter space $(\ell_{\text{tr}}, \ell_{\text{a}}, \mu)$. In this case, the P3 approximation was used to analyze experimental data.

Once transport parameters were identified, I used the P3 approximation for scattered and re-emitted light to analytically predict the spectrum of a white LED. This approach is described in Chapter 5. The spectrum of light can be easily converted to the color point. The conversion process is described in Appendix A. In this chapter, the analytical connection between the transport parameters of the phosphor and the color point of a light source was presented. This approach opens vast possibilities for the systematic design of white LEDs. A look-up table can be created where the phosphor type will be mapped to the area of colors it can generate. Application of the described approach will provide the design of the thickness of the phosphor layer d and the phosphor concentration ρ that will produce the desired color.

In Chapter 5 it was shown that design parameters strictly define the color point of the light source. I demonstrated in Chapter 7 that the color point of a white LED can be reversibly tuned without changing its design. This tuning is possible when the wavefront shaping technique is used to enhance the intensity at a preset wavelength. Theoretical predictions show that this technique works efficiently only for areas of illumination that are μm in size. In Chapter 7, it was demonstrated that enhancement reaches almost 1.5 for mm size areas of illumination. The enhancement of light at different wavelengths had different magnitude, which allowed reversible tuning of the color point.

I have developed tools that can drastically change the white LED design process, allowing analytical methods to be widely used. It will enable access to a large design parameter space by using an analytical algorithm without adjustable parameters. While my current solutions provide a one-dimensional description of the light propagation, the analytical solutions of the P3 approximation in the 3D geometry is already available. It describes the light propagation in complex geometries that are regularly employed in white LED design. My

approach increases the design efficiency by avoiding recurring design efforts and decrease the cost of ownership of white LEDs units for worldwide users.

Nederlandse samenvatting

Verlichting is een cruciale technologie die elke dag wordt gebruikt. De introductie van de witte lichtgevende diode (LED) dit is gebaseerd op de Nobelprijs-winnende technologie van een blauwe LED in combinatie met een fosforlaag, heeft het energieverbruik voor verlichting drastisch verminderd. Ondanks de sneltoenemende marktpenetratie, zijn er momenteel slechts een klein aantal LED ontwerpmethodes. In dit proefschrift beschrijf ik door mij ontwikkelde experimentele en theoretische hulpmiddelen die de eigenschappen van fosforpoeder analytisch verbinden met het kleurpunt van een witte LED. Fosfordeeltjes in de diffusorlaag verstrooien, absorberen en geven invallend licht opnieuw af. Absorptie en verstrooiing hebben een vergelijkbare invloed op het licht uitgezonden door een LED. Daarom kunnen geen van beide effecten worden verwaarloosd. Vanwege hoge absorptie kunnen veelgebruikte analytische hulpmiddelen de optische eigenschappen van fosfordiffusielagen niet beschrijven. In dit proefschrift stel ik experimentele en theoretische hulpmiddelen voor die met een nieuwe systematische benadering van lichtdiffusie en re-emissie in fosforlagen volledig beschrijven.

De P3-benadering is gekozen als het centrale theoretische raamwerk dat ons in staat stelt de lichtvoortplanting in verstrooiende materialen met een sterke absorptie te beschrijven. Deze benadering is afgeleid van de meer algemene stralingsoverdrachtsvergelijking (radiative transfer equation - RTE); gedetailleerde stappen van afleidingen worden beschreven in hoofdstuk 2. De vergelijkingen van de P3-benadering voor een parallelle plaat zijn analytisch oplosbaar en gepresenteerd aan het einde van hoofdstuk 2.

De P1-benadering is de eerste-orde benadering van RTE en heeft een veel eenvoudigere analytische oplossing dan de P3-benadering. In hoofdstuk 6 is aangetoond dat de P1-benadering een aanzienlijke fout geeft, van de orde van 30%, bij het schatten van transportparameters voor de verstrooiende materialen met sterke absorptie, en op sommige materialen zelf niet kan worden toegepast. Om het geldigheidsbereik voor P1-benadering te identificeren, leid ik analytische expressies af die foutschattingen van de P1-benadering voor het hele bereik van de transportparameters bieden. Deze schattingen worden gepresenteerd

in de vorm van 2D-kaarten.

Om gegevens te analyseren met de P1- en de P3-benaderingen, heb ik metingen uitgevoerd in het bereik waar verstrooid licht en opnieuw uitgestraald licht spectraal gescheiden worden. Conventionele meettechnieken leveren een dergelijke scheiding bij de bestudering van verstrooiende materialen met breedbandemissie niet op. In de hoofdstukken 3 en 4 presenteer ik een nieuwe experimentele opstelling die het probleem oplost. Deze opstelling maakt gebruik van smalband-verlichting, waardoor het verstoorde en opnieuw uitgestraalde licht altijd kan worden onderscheiden.

Het centrale doel van de hoofdstukken 3 en 4 is om de transportparameters van de fosforplaten te identificeren die typisch worden gebruikt voor witte LED's. In hoofdstuk 3 zijn de parameters $(\ell_{\text{tr}}, \ell_{\text{a}})$, en de P1-benadering gebruikt om deze parameters te extraheren. In Hoofdstuk 4 is een meer verfijnd experiment uitgevoerd en kon een bredere parameter-ruimte $(\ell_{\text{tr}}, \ell_{\text{a}}, \mu)$ worden geïdentificeerd. In dit geval is de P3-benadering gebruikt om experimentele gegevens te analyseren.

Nadat de transportparameters zijn geïdentificeerd, gebruik ik de P3-benadering voor verstrooid en opnieuw uitgezonden licht om het spectrum van een witte LED analytisch te voorspellen. Deze aanpak is beschreven in hoofdstuk 5. Het spectrum van het licht kan eenvoudig worden omgezet in het kleurpunt. Dit conversieproces wordt beschreven in Bijlage A. In dit hoofdstuk wordt de analytische verbinding tussen de transportparameters van de fosfor en het kleurpunt van een lichtbron gepresenteerd. Deze aanpak opent enorme mogelijkheden voor het systematisch ontwerpen van witte LED's. Er kan een opzoektabel worden gemaakt waarin het fosfortype wordt toegewezen aan het gebied met kleuren dat het kan genereren. Toepassing van de beschreven aanpak zal de dikte van de fosforlaag d en de fosforconcentratie ρ voorschrijven die de gewenste kleur zal produceren.

In hoofdstuk 5 wordt aangetoond dat de ontwerpparameters het kleurpunt van de lichtbron uniek bepalen. In hoofdstuk 7 heb ik aangetoond dat het kleurpunt van de witte LED omkeerbaar kan worden gevarieerd zonder het ontwerp te veranderen. Deze afstemming is mogelijk wanneer een golffrontaanpassingstechniek wordt gebruikt om de intensiteit bij een vooraf ingestelde golflengte te verbeteren. Theoretische voorspellingen hadden aangetoond dat golffrontaanpassing efficiënt werkt voor verlichtgebieden die μm groot zijn. In hoofdstuk 7 wordt echter aangetoond dat golffrontaanpassing leidt tot een verbetering met een factor van bijna 1.5 bij verlichtingsgebieden met een mm-grootte. De toename van het licht verschilt voor verschillende golflengtes, wat omkeerbare afstemming van het kleurpunt mogelijk maakte.

Ik heb theoretischemiddelen ontwikkeld die het witte LED-ontwerpproces drastisch

kunnen veranderen, waardoor analytische methoden op grote schaal kunnen worden gebruikt. Het zal een grote ontwerpparameterruimte mogelijk maken met behulp van een analytisch algoritme zonder aanpasbare parameters. Hoewel mijn huidige oplossingen een ééndimensionale beschrijving van de lichtvoortplanting bieden, zijn de analytische oplossingen van P3-benadering in 3D-geometrieën al beschikbaar. Ze kunnen lichtvoortplanting in complexe geometrieën beschrijven die regelmatig in witte LED-ontwerpen worden gebruikt. Mijn aanpak verhoogt de ontwerpefficiëntie door terugkerende ontwerpinspanningen te vermijden en de eigendomskosten van witte LED's voor wereldwijde gebruikers te verlagen.

Appendix A

Color point

A.1 What is color?

Color is a complicated concept that includes both a strict physical definition and human psychology - a rare combination. To demonstrate the psychological aspect of this problem, we will discuss the famous case of 'The dress.' On 26 February 2015 on the social service Tumblr a user posted a washed-out color photograph of a dress with hashtag #thedress, shown in Fig. A.1. Users were asked to answer a simple question: 'what is the color of the dress?' It appears that only 57% of the people perceived the dress as blue and black. An other 30% saw it as gold and white, and 2% saw it as something else [120].



The scientist suggested that the perception of colors on the photo depends on the white light source used to illuminate the object and how the brain processes this illumination [121]. The final answer to this question is a topic of ongoing scientific investigation in neuroscience and vision science. This example illustrates the psychological aspects of color perception and importance of a white light source characteristics in our lives.

Although color perception depends on the individual, there were formulated various methods to characterize color of objects, light sources, and their interaction [93]. These methods include rigorous characterization of the eye photodetector sensitivity. In this appendix, we describe how to identify the color perceived by a human

Figure A.1: **'The dress.'** The color of the dress is blue and black. It may appear white and golden. Figure adopted from [120]. Dress image reproduced with permission from Cecilia Bleasdale.

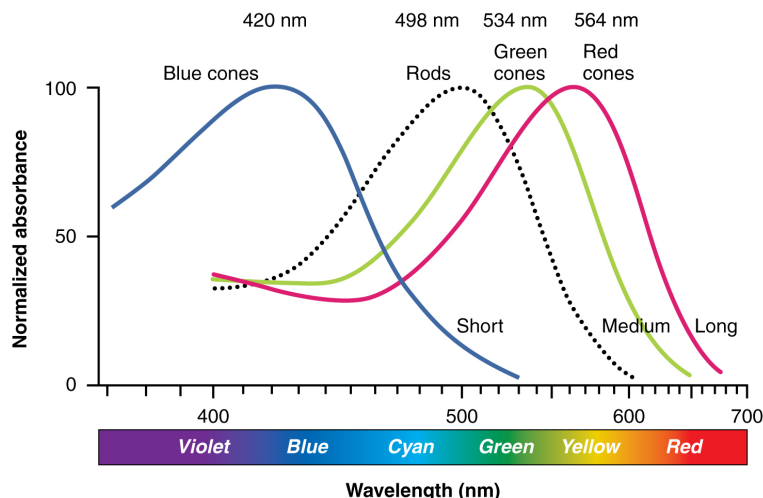


Figure A.2: **Absorbance curve of the eye sensors.** The sensitivity of rods and cones of the eye as a function of wavelength. Figure adopted from [122].

from the spectrum measured in the lab.

A.2 Human eye

The human eye contains two types of photodetectors: rods and cones [55]. Rods are the color insensitive type of photodetectors. They are optimized for night vision and hence sensitive to deficient light levels. The absorbance curve as a function of wavelength for the rods is shown in Fig. A.2. Cones are color sensitive photodetectors. There are three types of cones red, green and blue (see Fig. A.2). Every type of cone is optimized to be sensitive in its own part of the spectrum. The absorbance curves for all three types of detectors overlap, covering all visible spectrum. Cones perform well at high light levels and are mainly used during the day. There are about 120 million rods and 6 million cones inside the human eye.

The distribution of cones and rods is not uniform over the surface of the eye (see Fig. A.3). The cones are mainly concentrated in the center of the eye, and rods are concentrated on the periphery. Cones have a higher spatial resolution compared to rods. There are no photoreceptors at an angle of 20 degrees from the fovea of the eye. That is the position where the eye nerve is connected to the eye, and this area is called the blind spot.

A.3 From spectrum to color

The visible spectrum is a range of electromagnetic radiation that can be registered by our eye [55]. Each wavelength in the visible spectrum corresponds to the spectrally pure

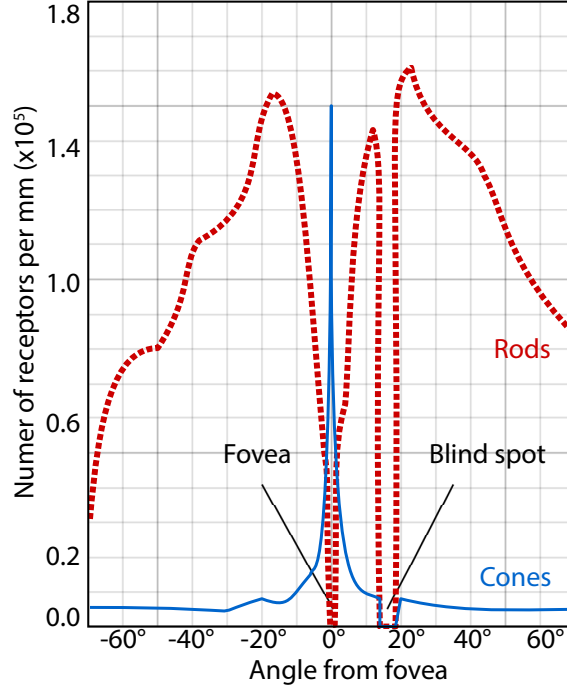


Figure A.3: **The density of rods and cones in the eye.** The distribution of cones and rods inside the eye as a function of the angle from the fovea. Figure adopted from [123].

monochromatic color seen by the human. The spectrally pure colors are not restricted to the variety of colors in nature. The purple color is a combination of blue and red spectrally pure colors. Orange is a spectrally pure color (600 nm), it can also be produced by mixing red (700 nm) and yellow (580 nm). The eye cannot detect the two spectral components that resemble orange color, so we say that the eye is a synthesizer. On the contrary, the ear is an analyzer, because when we listen to an orchestra, separate instruments can be heard.

Colors can be described using three parameters. Here we use hue, saturation, and luminance (see Fig. A.4). Saturation characterizes the amount of white color added to the spectrally pure color. For example, if we add white to red color, we will obtain the pink color. The saturation is 100% if no white is added. To define luminance, we consider the following examples. If two objects with the identical color are illuminated differently (one object is in the darkness, and another one is strongly illuminated), they appear to have a different color. This is the essence of the luminance parameter.

The CIE 1931 color space is the first quantitative method to convert color to spectrum and *vice versa*. One of the most widely known color space is the RGB color space, which stands for red, green and blue color space. To define the main quantities of this space the color matching experiments was done (see Fig. A.5). An observer looks at the screen that is divided into two parts as shown in Fig. A.5(a). One part of the screen is illuminated

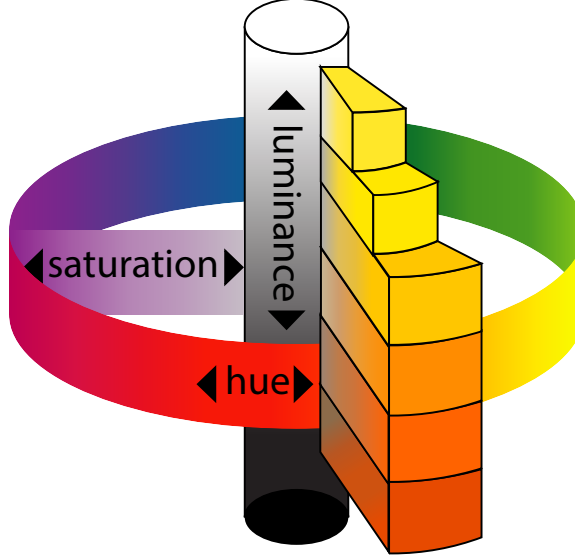


Figure A.4: **Hue, saturation and luminance diagram.** Hue changes on the circumference of the circle. Saturation changes along the diameter of the circle. Luminance changes on the axes that cross the center of the circle perpendicularly.

with three spectrally pure colors (red, green and blue). Another part is illuminated with arbitrary color. The observer is asked to match the color of the two screens by adjusting the power of the three light sources. When the colors have matched, the value of the individual power settings is recorded as a function of wavelength (see Fig. A.5). When the color of the screens cannot be matched exactly then the additional light source is used to illuminate the reference screen. The power of the additional source is recorded as negative power. In Fig. A.5(b) we show functions $\bar{x}$, $\bar{y}$ and $\bar{z}$, and they are already normalized in such a way that no negative values are present.

To obtain the RGB coordinates of the measured spectrum the following mathematical procedures should be performed. First we calculate the tristimulus values of the spectrum

$$X = k \sum_i \Phi(\lambda_i) \bar{x}(\lambda_i) \Delta \lambda_i, \quad (\text{A.1})$$

$$Y = k \sum_i \Phi(\lambda_i) \bar{y}(\lambda_i) \Delta \lambda_i, \quad (\text{A.2})$$

$$Z = k \sum_i \Phi(\lambda_i) \bar{z}(\lambda_i) \Delta \lambda_i, \quad (\text{A.3})$$

where k is the normalization constant that often chosen in such way that $Y = 100$ for the white color. The procedure of calculating the tristimulus value is shown in Fig. A.6. XYZ can be easily converted to RGB using a conversion matrix. The conversion matrix depends on the choice of the chosen white color spectrum. In this thesis we use white color D65

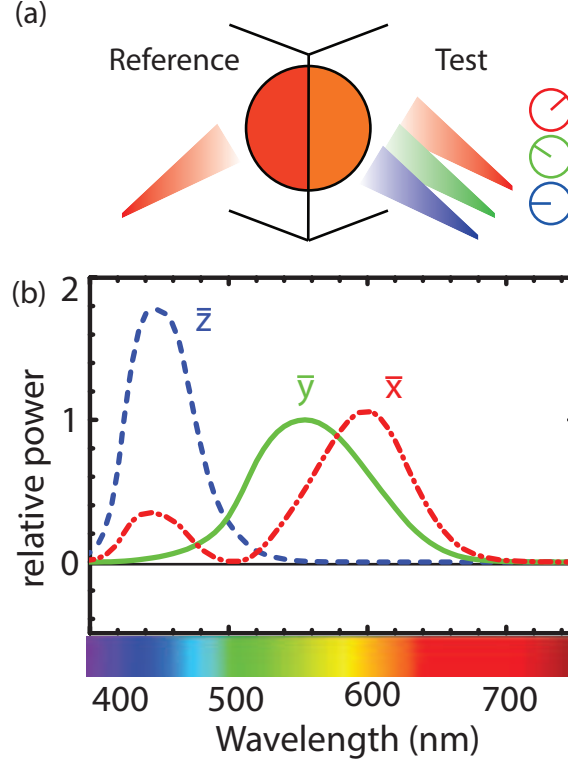


Figure A.5: **Color matching experiment.** (a) The color matching experimental setup. The color is set on the left part of the screen - the reference screen. Knobs on the right side of the screen are adjusted to match the color of the test screen to the reference screen. (b) The color matching functions $\bar{x}$, $\bar{y}$ and $\bar{z}$.

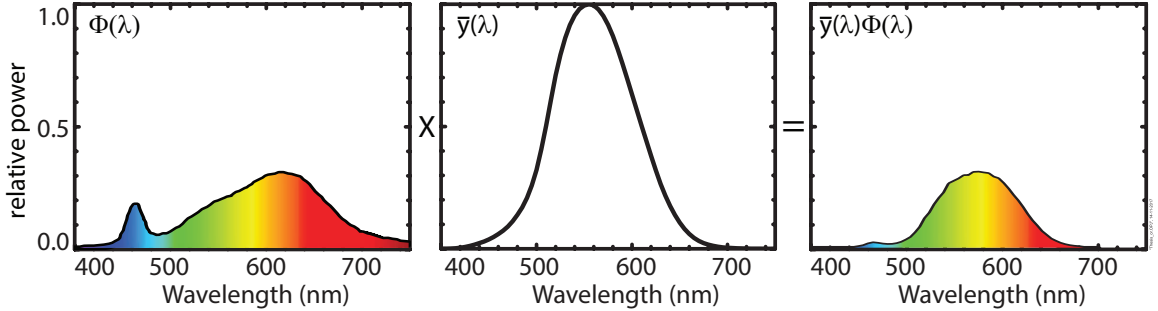


Figure A.6: **From color space $\bar{y}$ to color space Y .** The light source with the spectral density $\Phi(\lambda_i)$ is multiplied by the color matching function of the eye $\bar{y}(\lambda_i)$ to obtain the stimulus registered by the human eye $\bar{y}(\lambda_i)\Phi(\lambda_i)$. When we sum $\bar{y}(\lambda_i)\Phi(\lambda_i)$ over all i 's in the visible spectrum we receive Y that is the area under the curve in the last figure.

shown in Fig. A.7. $D65$ roughly represents the average midday light in Western Europe. It is a combination of the sunlight spectrum and light diffused by the atmosphere. We chose $Y = 100$ to define $k = 1.1149$, as mentioned above.

In this thesis we used chromaticity values to indicate the color point on the color space.

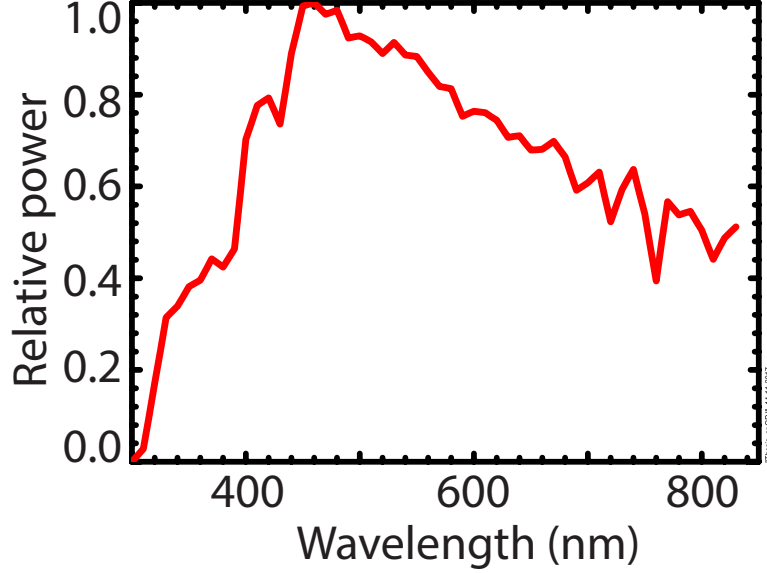


Figure A.7: **Spectrum of the white color D65.** The chromaticity values of the *D65* are $(x, y) = (0.3125, 0.3290)$.

Chromaticity values xyz are calculated as follows

$$x = \frac{X}{X+Y+Z}, \quad (\text{A.4})$$

$$y = \frac{Y}{X+Y+Z}, \quad (\text{A.5})$$

$$z = 1 - x - y. \quad (\text{A.6})$$

These values are used as coordinates of the color point in the color space.

A.4 Color of objects

The color of an object depends on two factors: the reflectance of the object as a function of wavelength and the power spectrum of the light source that illuminates the object. In this section, we do not consider the psychological aspects described in Sec. A.1. In Fig. A.8 we show how the spectrum of the illumination influences the color perceived by the eye. The red object with reflectance spectrum shown in Fig. A.8(b) is illuminated by a RGB LED with the power spectrum shown in Fig. A.8(a). The stimulus at the eye is calculated by multiplying the power spectrum of the source by the reflectance spectrum of the object. The result of the multiplication is shown in Fig. A.8(c). We converted the spectra shown in Fig. A.8(b-c) to the color perceived by the human eye. The results of the conversion are shown in the inset of Fig. A.8(b). The color of the middle colored square indicated with

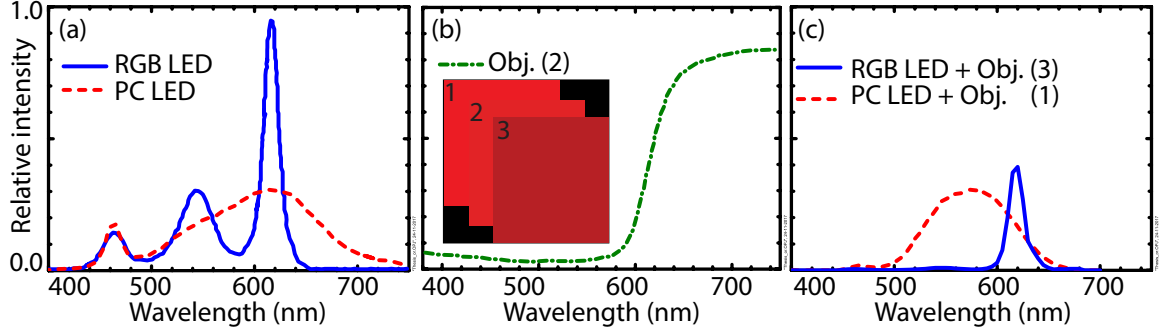


Figure A.8: **Color vs illumination conditions.** (a) Spectra of two white light sources that illuminate a test object. (b) The reflectance spectrum of the test object. The inset shows three squares with different shades of the red color. These colors are the result of the conversion of spectra from panels (b-c). Each color is indicated with the number that corresponds to the spectrum number mentioned in the legend of panels (b-c). (c) The stimulus registered by the eye.

number 2 represents the real color of the object. Squares 1 and 3 represent color perceived by the eye when illuminated with two different light sources. We see that the shape of the spectrum of the object is radically different from the spectra perceived by the eye. However, the resulting colors are very similar. Colors 1 and 2 can hardly be distinguished.

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