FINNISH METEOROLOGICAL INSTITUTE
CONTRIBUTIONS

No. 37

LIGHT SCATTERING BY NONSPHERICAL ATMOSPHERIC PARTICLES

Timo Nousiainen

Division of Atmospheric Sciences
Department of Physical Sciences
Faculty of Science
University of Helsinki
Helsinki, Finland

ACADEMIC DISSERTATION in meteorology

To be presented, with the permission of the Faculty of Science of the University of Helsinki, for public critisism in Auditorium Porthania IV (Yliopistonkatu 3) on June 1st, 2002, at 10 a.m.

Finnish Meteorological Institute
Helsinki, 2002

ISBN 951-697-560-7 (print)
ISBN 952-10-0584-X (pdf)
ISSN 0782-6117

Yliopistopaino
Helsinki, 2002

Published by Finnish Meteorological Institute
P.O. Box 503
FIN-00101 Helsinki
Finland

Name and number of publication
Contributions 37, FMI-CONT-37

Date
May 2002

Authors
Timo Nousiainen

Name of project	Commissioned by

Title
Light scattering by nonspherical atmospheric particles

Abstract

Scattering is a very common and important phenomenon and needs to be taken into account in many meteorological applications from remote sensing to climate modeling. In this thesis, a representative set of different meteorological scattering problems has been solved using state-of-the-art single-scattering models in order to study the importance of sophisticated single-scattering modeling for meteorological applications. The main focus is on the detailed shape modeling of scatterers. As atmospheric scatterers (hydrometeors, aerosol particles) have generally irregular and unique shapes - with the notable exception of small liquid droplets which are spherical - statistical shape modeling is vastly exercised.

The scattering studies are focused on particles either smaller or much larger than the wavelength of light, as there is presently no method which could handle detailed, realistic particle shapes in a region between these extremes. The studied cases include scattering of visible light by oscillating raindrops, scattering of microwaves by large ice particles (graupel, hail), and scattering of visible light by large mineral dust particles.

It is shown how the importance of shape on scattering depends on the size parameter: for small size parameters, it suffices to model the particle elongation properly, and even that is necessary only for polarization quantities; for large size parameters, it is not sufficient to model the global shape in detail, but also the surface texture needs to be taken into account properly. Internal structure of particles is shown to affect scattering strongly and generally should be taken into account. In addition, a new rainbow phenomenon is found, and a general shape model for natural irregular particles is suggested. The work has also resulted in an industrial application (precipitation type sensor).

In general, the results demonstrate the importance of sophisticated scattering modeling, allow a qualitative evaluation of the relative importance of different factors affecting scattering, and illustrate different error sources and typical difficulties in scattering modeling. Finally, some general quidelines are given for accurate single-scattering modeling.

Publishing unit
Geophysical Research

Classification (UDC)	Key words
551.521.3, 551.593, 537.874, 551.510.42	light scattering, absorption, polarization, atmosphere, particles, aerosol, nonspherical shape, modeling, remote sensing

ISSN and key name	ISBN
0782-6117 Finnish Meteorological Institute Contributions	951-697-560-7 (print); 952-10-0584-X (pdf)

Language	Pages	Price
English	180	

Sold by	Note
Finnish Meteorological Institute Library P.O. Box 503 FIN-00101 Helsinki, Finland	

Julkaisija	Ilmatieteen laitos PL 503 00101 HELSINKI	Julkaisun sarja, numero ja raporttikoodi Contributions 37, FMI-CONT-37 Julkaisuaika Toukokuu 2002

Tekijä(t)
Timo Nousiainen

Projektin nimi	Toimeksiantaja

Nimeke
Valonsironta ilmakehän ei-pallomaisista hiukkasista

Tiivistelmä

Sironta on hyvin yleinen ja tärkeä ilmiö, joka täytyy huomioida monissa meteorologisissa sovellutuksissa kaukokartoituksesta ilmastomallitukseen. Tässä väitöskirjassa on tutkittu tarkan sirontamallituksen tarpeellisuutta erilaisissa meteorologisissa sovellutuksissa ratkaisemalla huipputasoa olevilla sirontamalleilla joukko erilaisia meteorologisia sirontaongelmia. Huomiota on kiinnitetty erityisesti tarkan muotomallituksen tärkeyteen. Koska ilmakehässä olevat sirottajat ovat yleensä muodoltaan epäsäännöllisiä mutta kuitenkin yksilöllisiä - merkittävänä poikkeuksena pienet nestepisarat jotka ovat pallomaisia - muotomallitus on tehty pääsääntöisesti tilastollisesti.

Tarkastelut keskittyvät lähinnä hiukkasiin jotka ovat joko aallonpituutta pienempiä tai selvästi sitä suurempia, sillä väliinjäävässä alueessa toimivia realistisen muotomallituksen mahdollistavia sirontamalleja ei ole toistaiseksi tarjolla. Tutkittuja tapauksia ovat näkyvän valon sironta värähtelevistä sadepisaroista, mikroaaltojen sironta suurista jäähiukkasista (lumirakeet, rakeet) sekä näkyvän valon sironta suurista mineraalipölyhiukkasista.

Muotomallituksen tärkeyden osoitetaan riippuvan sirottajien kokoparametrista: pienillä kokoparametrin arvoilla riittää mallintaa hiukkasten pitkulaisuus oikein ja sekin on tarpeen vain polarisaatiosuureiden osalta. Toisaalta suurilla kokoparametrin arvoilla ei riitä mallintaa hiukkasen globaalia muoto yksityiskohtaisesti, vaan myös hiukkasen pinnan hienorakenne täytyy ottaa huomioon. Myös hiukkasten sisäisen rakenteen osoitetaan vaikuttavan sirontaan niin merkittävästi että se tulisi yleensä ottaa huomioon. Tutkimuksen sivutuotteena on lisäksi löydetty uusi sateenkaari-ilmiö, ja luonnollisille mineraalipölyhiukkasille ehdotetaan yleistä muotomallia. Työ on myös edesauttanut teollisen sovellutuksen (optinen sadetyypin tunnistin) synnyssä.

Yleensä ottaen tulokset havainnollistavat tarkan sirontamallituksen tärkeyttä, mahdollistavat eri tekijöiden suhteellisen tärkeyden kvalitatiivisen arvioinnin erilaisissa sirontaongelmissa, sekä tuovat esiin erilaisia virhelähteitä ja tyypillisiä sirontamallitukseen liittyviä ongelmia. Väitöskirja sisältää myös yleisiä ohjeita tarkalle sirontamallitukselle.

Julkaisijayksikkö
Geofysiikka

Luokitus (UDK) 551.521.3, 551.593, 537.874, 551.510.42	Asiasanat valon sironta, absorptio, polarisaatio, ilmakehä, hiukkaset, aerosoli, ei-pallomainen muoto, mallitus, kaukokartoitus

ISSN ja avainnimeke 0782-6117 Finnish Meteorological Institute Contributions	ISBN 951-697-560-7 (print); 952-10-0584-X (pdf)

Kieli englanti	Sivumäärä 180	Hinta

Myynti Ilmatieteen laitos, kirjasto PL 503, 00101 HELSINKI	Lisätietoja

Preface

Working in three different organizations during my graduate years, I have met very many scientists and other personnel who have influenced my work. It would be hopeless to name them all here, yet they deserve my thanks. So, I begin my thanks by thanking all the anonymous colleagues who have interacted with me during these years. My colleagues at the Observatory, where I was initiated to the "secrets" of light scattering, provided me with years (two years, four months) that I will never forget. During my years at the Department of Meteorology, where I started using my new skills to meteorological applications, I was shown what it is like to be a scientist (as well as what it is not). My time at the Geophysical Research is short still, but I have already become addicted to its nearly magical atmosphere and research environment, found the benefits of the round balcony, and increased significantly my skill to play sähly.

I would like to specially thank my parents Anja and Jouko for all the love and support they have provided me with. I would also like to thank Kari Lumme for introducing me to the light scattering business and advising me during the years in the Observatory; Jouni Peltoniemi for always finding time to answer my (rather numerous) questions about light scattering, the universe, and everything; the members of Geodynamo (as well as the audience) for letting me play guitar; Niilo Siljamo for general LaTeX-related support during (and after) my years in the Department of Meteorology; and all those persons who, directly or indirectly, have contributed to the research contained in this thesis. Be it criticism, collaboration, or proof-reading, it is much appreciated. I am also in depth to my superiors at GEO who have allowed me to concentrate on the finishing off of my thesis. In addition, I wish to thank Vaisala Oyj for partial funding of the research carried out for Paper II of this thesis; Nordic Council of Ministers for funding my participation at the international light scattering conference in Helsinki, June 9-11, 1997; and the Chancellor of the University of Helsinki for providing me a travel grant to participate the 5th Conference on Light Scattering by Nonspherical Particles in Halifax, August 28 - September 1, 2000.

Last but definitely not least, I would like to thank my advisor Karri Muinonen, who has been patient and helpful both as an advisor and a collaborator and provided me with the theoretical support necessary for the successful completion of this work.

Summary of the thesis

This thesis consists of work carried out at the Observatory, University of Helsinki, at the Department of Meteorology, University of Helsinki (at present: Division of Atmospheric Sciences, Department of Physical Sciences), and at the division of Geophysical Research, Finnish Meteorological Institute. The main theme of the research is the importance of sophisticated single-scattering modeling, especially the importance of realistic particle shape, in meteorological scattering problems. Partially due to the restrictions of available single-scattering models, the work has not been focused on the meteorologically most important scattering processes. However, it does include a representative set of different kind of scattering problems, so that the results and conclusions might be generalized to other relevant scattering problems. The papers included consider scattering problems that have not been solved previously and present many new results that should be interesting also to scientists working outside meteorological scattering applications.

This thesis consists of an Introduction and five original papers:

Paper I: Muinonen, K., Nousiainen, T., Fast, P., Lumme, K., and Peltoniemi, J. I., 1996: Light scattering by Gaussian random particles: ray optics approximation, *J. Quant. Spectrosc. Radiat. Transfer*, **55**, 577–601.

Paper II: Nousiainen, T. and Muinonen, K., 1999: Light scattering by Gaussian, randomly oscillating raindrops, *J. Quant. Spectrosc. Radiat. Transfer*, **63**, 643–666.

Paper III: Nousiainen, T., 2000: Scattering of light by raindrops with single-mode oscillations, *J. Atmos. Sci.*, **57**, 789–802.

Paper IV: Nousiainen, T., Muinonen, K., Avelin, J., and Sihvola, A., 2001: Microwave backscattering by nonspherical ice particles at 5.6 GHz using second-order perturbation series, *J. Quant. Spectrosc. Radiat. Transfer*, **70**, 639–661.

Paper V: Nousiainen, T., Muinonen, K., and Räisänen, P., 2002: Scattering of light by large Saharan dust particles in a modified ray-optics approximation, *J. Geophys. Res.* (in press).

In Paper I, light scattering by so-called Gaussian random particles was studied systematically in the ray-optics approximation, varying both the particle shape statistics and the refractive index. This paper is the main publication of Gaussian random spheres, introducing the general spherical-harmonics formalism for well-defined Gaussian particles into light-scattering modeling. The author's contribution was to test and debug the scattering model, carry out the scattering simulations, and analyze and compile the results.

In Paper II, the importance of accurate shape modeling of large oscillating raindrops were studied. The oscillations were modeled using the Gaussian random sphere model, as the information about oscillations were considered insufficient for a deterministic model. Light scattering properties of oscillating raindrops were studied systematically for two distinct sizes by varying the oscillation statistics. Apparently for the first time, full 4π scattering matrix analysis was performed. Most of the work involved was carried out by the author; the original scattering model for Gaussian random spheres (not modified for raindrop geometries) was written by Karri Muinonen.

In Paper III, the study of light scattering by oscillating raindrops was continued. The random oscillation scheme was replaced by more physical statistical single-mode spherical harmonics scheme consistent with oscillation observations. Light scattering properties of raindrops with single-mode oscillations were systematically studied varying the angle of incidence, oscillation modes and amplitudes, drop sizes, and size distributions. For appropriate parts, the results were compared with those given in Paper II. To the author's knowledge, this work presents by far the most sophisticated scattering modeling of natural raindrops to date. Except for a few details, all the work was carried out by the author.

In Paper IV, microwave backscattering by nearly-spherical inhomogeneous ice particles was studied using a second-order perturbation series approximation adapted for Gaussian random spheres. De- and co-polarized backscattering cross sections were computed for varying particle geometry, composition, and internal structure. In this paper the radar backscattering is, apparently for the first time, studied using statistically given nonspherical shapes resembling natural particles. Most of the work was carried out by the author; the original scattering model was provided by Karri Muinonen, and effective refractive indices for different mixtures were provided by Juha Avelin and Ari Sihvola.

In Paper V, light scattering properties of Saharan mineral particles large compared to the wavelength were studied both in a traditional and in a modified ray-optics approximation. Particle shapes were based on a shape analysis of a sample of real Saharan particles, and model results were compared with scattering measurements carried out by Volten et al. (2001) using the same sample. This paper proposes an *ad hoc* simple Lambertian modification to traditional ray-optics approximation to take into account the small-scale surface structure and internal inhomogeneity of mineral particles; this modification allows, for the first time, good agreement between simulated and measured scattering properties for natural mineral particles using realistic model particle shapes. Apart from the Lambertian modification by Karri Muinonen, and the radiative transfer contribution by Petri Räisänen, all the work was carried out by the author.

Contents

1 Introduction

Scattering of electromagnetic (EM) radiation, hereafter light scattering, is a manifestation of the interaction between electromagnetic radiation and matter. If a photon interacts with a scattering medium without interchanging energy, we have elastic scattering with the scattered frequency equal to the incident frequency. If the photon energy is changed in the process, we have inelastic scattering with the scattered frequency not equal to the incident frequency. This thesis is confined to elastic scattering, with the exception of absorption that needs to be taken into account and is actually a special case of inelastic scattering. The EM radiation is assumed to consist of plane waves, with the notion that an arbitrary radiation field can be expressed as a suitable superposition of plane waves (e.g., Bohren and Huffman, 1983).

The classical interpretation of elastic scattering is the following: a time-dependent electric field associated to EM radiation forces charged elementary particles (mainly electrons) within matter to oscillate at its frequency. The oscillating charges are in accelerating motion and thus they radiate electromagnetic radiation (e.g., Bohren and Huffman, 1983). Scattered light, which is distributed in all directions with an angular dependence characteristic to the scattering event, consists of this re-radiated energy and originates from the energy of the incident radiation. Absorption can then be considered the part of the incident energy that is lost from the incident radiation but is not re-radiated as scattered energy. This energy is stored in the scattering medium as internal energy and is emitted with a spectrum of wavelengths depending on, e.g., the temperature and the material of the particle (e.g., Liou, 1980). In most meteorological applications, the wavelengths of scattered and emitted light do not overlap significantly.

There are several reasons why scattering is an important phenomenon to study. First, it is a very common phenomenon. All matter scatters light, and indeed, most things are visible to us only because they scatter light. There are only few objects, like the Sun, a candle flame, and red-hot lava that emit visible light sufficiently to be seen. Second, the properties of the scattered light depend on the physical properties of the scatterer, and this information is carried to an observer at the speed of light. This allows remote sensing of the physical properties of distant targets. Third, scattering affects the way radiation propagates in a medium. It is often of fundamental importance to take scattering into account in radiative transfer computations. Such computations are needed in a wide range of applications from engineering to climate modeling.

Although the physics of scattering is well understood in principle, there is no universal solution to a scattering problem — the resulting set of equations cannot be solved analytically except in few simple cases. Numerically the solution can be found, but this is practical only in some cases (when scatterers are sufficiently small compared with the wavelength). Thus, it has been necessary to develop different methods, most of which are approximations, for different kind of scattering problems. Indeed, there are quite a few different methods available today, out of which it is often a problem itself to choose the one most appropriate for the scattering problem at hand. As a consequence, ideal methods are not always used.

Today, the development of scattering methods is mostly confined in developing better numerical techniques. On one hand, faster and more general scattering models are being developed. On the other hand, methods to solve new problems and take more details into account are looked for. Less effort is put into the development of analytical methods, and it is unlikely that the number of special cases that can be handled analytically will increase considerably in the future (Mishchenko et al., 2000b). This thesis is largely based on applying existing methods. The model development involved is largely confined to modifying the models for new geometries and to automate certain procedures such as an integration over a size distribution. However, the models used are state-of-the-art products and, in case of Papers IV and V, used for the first time for a real scattering problem.

In the thesis, several meteorological scattering problems have been solved using sophisticated scattering methods, with two main goals in mind: first, to establish the effect and the relative importance of different physical properties of atmospheric scatterers on scattering; and second, to consider the possible errors caused by the use of more simplified methods. It is hoped that this thesis would provide some insight to factors considered important in different scattering problems and would thus help readers in choosing a proper scattering method for their applications. In addition to providing guidance to scientists who apply scattering methods in their work but are not necessarily deeply involved in the scattering studies, it is hoped that the work would also be of interest to people specialized in scattering.

Three different meteorological scattering problems are considered in detail: scattering of visible light by oscillating raindrops (Papers II and III), scattering of microwaves by nearly-spherical ice particles (Paper IV), and scattering of visible light by large mineral dust particles (Paper V). In the following, a short summary is given of the relation of these studies to the previous works. More thorough discussions are given in the appropriate papers.

Paper I is not focused on any particular scattering problem imposed by nature, but rather on the general features of scattering by irregular particles large compared to the wavelength. In many ways, it also provides a foundation for this thesis. It introduces the statistical random shape model called the Gaussian random sphere, which is applied in all papers included in this thesis except in Paper III. This statistical shape model for nonspherical particles originates from the stochastically rough particle shape model by Peltoniemi et al. (1989) and was later further developed to the Gaussian random sphere model by Muinonen (1996, 1998, 2000b). For a recent review of the history and the present state of light scattering modeling using stochastically shaped scatterers, see Muinonen (2000b). In addition, Paper I introduces the Monte Carlo ray tracing geometric optics algorithm, which was used, after appropriate modifications, in Papers II and III. This algorithm performs ray tracing in a matrix form, so the full scattering matrix can be solved simultaneously without the need to vary the polarization state of the incident light. The entire ray tree is solved, differently to the Markovian approach adapted by, e.g., Peltoniemi et al. (1989). Finally, the implementation of the Kirchhoff approximation (e.g., Jackson, 1975; Muinonen et al., 1989) for diffraction is introduced in Paper I and used also in Paper V.

The study of light scattering by oscillating raindrops, carried out in Papers II and III, is much benefited from the fact that the exact shapes, orientations, and size distributions of falling raindrops are widely studied topics (see, e.g., Hendry et al., 1976; Beard and Chuang, 1987; Beard et al., 1989; Pruppacher and Klett, 1997, and references therein). Perhaps the most important application for this information is the rainfall measurement by dual-polarized radars, which allow a comparison of backscattering by raindrops for horizontally and vertically polarized radar beams (see, e.g., Goddard et al., 1982; Sauvageot, 1994). A good spatial and temporal resolution of the rainfall rate can be measured reasonably only by using radars, but so far the radar-measured absolute rainfall rates are not sufficiently accurate for many practical applications. However, the knowledge of raindrop shapes and orientations, especially as a function of drop size, together with the use of dual-polarized radars, can increase this accuracy considerably (e.g., Oguchi, 1983; Herzegh and Jameson, 1992). An accurate measurement of rainfall rate would be of great value, even if it was limited to the vicinity of the radar where the radar beam is near the ground level. In addition, the same information is beneficial also for optical applications. For example, the scattering model used in Paper II has also been used in developing a new optical precipitation type sensor at Vaisala Oyj (Lehtelä et al., 1999). Such an instrument can be used, e.g., to detect the presence of liquid water drops in subzero temperatures, a situation hazardous to, e.g., traffic and power lines.

Natural raindrops do not have tear-like shapes, contrary to popular belief. Rather, small raindrops are practically spherical, while larger drops are increasingly flattened from the bottom side, resembling somewhat to an oblate spheroidal shape (e.g., Beard et al., 1989; Pruppacher and Klett, 1997). In addition, large raindrops tend to oscillate (e.g., Jones, 1959; Tokay and Beard, 1996). These oscillations consist, according to both observations and theoretical considerations, of varying spherical harmonics modes (see, e.g., Trinh et al., 1982; Kubesh and Beard, 1993, and references therein). Interestingly, the physics of the oscillations of water drops (and jets) have been studied already in the 19th century (e.g., Rayleigh, 1879). The orientation of falling raindrops can be considered horizontal with small deviations (see, e.g., Beard and Jameson, 1983; Pruppacher and Klett, 1997, and references therein), following from the aerodynamic forcing. Raindrop size distributions vary, but temporally or spatially averaged distributions resemble the Marshall-Palmer type negative-exponential size distribution (Marshall and Palmer, 1948), while "instantaneous" distributions tend to be more monodisperse (Joss and Gori, 1978). For a thorough review of raindrop geometry, see, e.g., Nousiainen (1997) or Pruppacher and Klett (1997).

Microwave backscattering by large atmospheric ice particles, considered in Paper IV, is also a widely studied topic. Most operational radars apply the Rayleigh theory as a basis for the so-called radar equation which connects the backscattered signal to the total mass of scatterers. However, for typical weather radars with a wavelength of about 5 cm, the assumptions of the Rayleigh theory (see, e.g., Bohren and Huffman, 1983) are not satisfied in the case of large ice particles, especially if the particles are also partially melted. There have been many studies using more accurate methods (see, e.g., Oguchi, 1983; Aydin, 2000; Haferman, 2000, and references therein), but these are largely confined to simplified shapes which have limited applicability especially when the (de)polarization quantities are studied. Yet, these quantities include additional independent information that can be taken advantage of by modern dual-polarized radars.

Natural ice particles impose a challenging light scattering problem. In Paper IV, only nearly-spherical (e.g., the deviation from spherical shape can be considered a minor perturbation) ice particles are studied, including—conforming to the classification given in Pruppacher and Klett (1997)—graupel and small-hail particles, sleet, and hailstones. These particles are typically rounded, irregular, or conical in shape (see, e.g., Matson and Huggins, 1980), and are composed of ice with varying amounts of air and liquid water trapped inside them (e.g., Chýlek et al., 1984). Melting particles often also have a liquid water coating. Fortunately, the inhomogeneity can be approximately taken into consideration with a straightforward way by applying a so-called effective-medium approximation (see, e.g., Sihvola, 1989; Chýlek et al., 2000). Falling hydrometeors generally prefer orientations maximizing drag, but for the nearly-spherical particles drag is almost independent of the orientation and thus the particles of interest here can be considered, as a first approximation, randomly oriented. The research carried out in Paper IV is different from the previous studies in the sense that truly irregular and yet well-defined particle shapes are used, allowing different and well-controlled study of the effect of particle shapes on scattering. This is especially relevant for depolarization.

Scattering of visible light by large mineral dust particles is studied in Paper V. Solving single-scattering properties of atmospheric mineral aerosol particles is a very relevant and contemporary scattering problem: they form the most predominant aerosol class observed from space, they have considerable impact on the radiative balance of the atmosphere, and they must be taken into account when atmospheric constituents, e.g. ozone, are monitored from space. At the same time, the single-scattering properties of mineral particles are needed in many other applications, such as in studies of Martian atmosphere or modeling scattering properties of particulate regolith. However, natural mineral particles provide a very challenging single-scattering problem, especially at visible light or wavelengths shorter than that. They are irregularly shaped, rather rounded particles with surface covered with structures in many scales. They are also inhomogeneous, and the size scale of inhomogeneities is generally too large to be taken into account by effective-medium approximations at visible light or wavelengths shorter than that. In addition, their typical size variation is so large that there is no single-scattering method available today which could handle it wholly and accurately. Indeed, only a so-called Lorenz-Mie theory could handle such a size range, but it is restricted to isotropic, homogeneous spheres. An accurate solution can be looked for by applying several different methods for different size ranges, but there are problems in this approach too: different methods make different assumptions and it may not be simple to combine them together; in addition, there is currently no practical and accurate method available for mineral particles slightly larger than the wavelength.

Traditionally, mineral aerosol particles have been assumed isotropic, homogeneous spheres. Indeed, even modern applications such as the OPAC database (Hess et al., 1998) or the AERONET network (Dubovik et al., 2002) adapt this assumption, and most, if not all, retrieval algorithms of satellite applications are based on similar assumptions. Considering the difficulty of an accurate modeling, it is not surprising that operational products rely on the Lorenz-Mie theory, which is fast, exact, and can handle the whole size distribution of particles. Nevertheless, it is a well-established fact that scattering by nonspherical particles is generally different to that of spherical particles (e.g., Mugnai and Wiscombe, 1980; Jaggard et al., 1981; Bohren and Huffman, 1983). Lately, spheroidal shapes have been used to evaluate the errors in satellite-borne retrieval of aerosol properties caused by the assumption of spherical shapes (Mishchenko et al., 1995, 1997; Pilinis and Li, 1998). Although these studies show major differences in scattering by spherical and spheroidal aerosol particles, they are not complete, as the effects of internal inhomogeneity and rough particle surface are not accounted for. Thus, in Paper V the importance of different factors on light scattering by natural mineral particles is studied taking into account also surface roughness and inhomogeneity. Such a study is largely made possible by the availability of laboratory measurements of natural mineral particle scattering (Volten et al., 2001; Muñoz et al., 2001) which can be used as a benchmark in the absence of an exact theoretical solution. The research carried out in Paper V is different with other papers included in this thesis also in that respect that shape modeling is not based on literature, but instead, a shape analysis is part of the study.

The structure of this thesis is the following: In Chapter 2, theoretical aspects of light scattering are considered. The basic physical characteristics, mathematical tools, and basic definitions are discussed in Section 2.1. Section 2.2 considers in further detail the geometrical aspects important for scattering. Different light scattering methods are explained in Section 2.3, concentrating on the methods applied in this thesis. Chapter 3 presents the main results of the papers included, with Section 3.1 dedicated to scattering of visible light by raindrops, Section 3.2 to microwave backscattering by nearly-spherical ice particles, and Section 3.3 to scattering of visible light by mineral aerosol particles. The summary and conclusions are given in Chapter 4. Finally, after the list of References and a short Errata, Papers I–V follow.

2 Theoretical concepts

2.1 Physical background and definitions

Scattering by a single particle is customarily called single scattering, and the scattering properties of a particle single-scattering properties. These properties define the single-scattering process, i.e., relate the incident and the scattered light. As opposed to single scattering, multiple scattering consists of sequential single-scattering processes. Multiple-scattering problems are always radiative transfer problems and are not given much weight in this thesis. From a single-scattering point of view, it makes no difference whether the incident radiation is emitted or light that has already been scattered (van de Hulst, 1981).

Light scattering by a single particle depends on the properties of the incident radiation, the properties of the material the particle is composed of, and the geometry of the particle and the scattering process. The most important properties of incident radiation are the wavelength and the state of polarization. The amplitude (intensity) of the radiation is of secondary importance (as long as it is sufficiently low not to affect the properties of the scatterer or the medium significantly), as it does not affect the angle or the wavelength dependence of scattering. There are many ways to describe incident and scattered EM fields, the choice of which affects the mathematical formalism for the scattering process.

The most important characteristic of the matter is the complex (absolute) refractive index $N = n_r + in_i$ which depends on the polarizability and the number density of molecules. The refractive index is connected to the electric and magnetic properties of matter by

$$N = \sqrt{\frac{\varepsilon\mu}{\varepsilon_0\mu_0}}, \tag{2.1}$$

where ε and ε_0 are the (complex) permittivities of the matter and free space, respectively, and μ and μ_0 the corresponding (complex) permeabilities. Usually, within a sufficient accuracy, $N = \sqrt{\varepsilon/\varepsilon_0} = \sqrt{\varepsilon_r}$ (Jackson, 1975).

N is a function of the wavelength, and the composition and the density of the material. Its real part is related to the phase velocity of light in the material, while the imaginary part describes the absorption of light by the material (e.g., Bohren and Huffman, 1983). As absorption and scattering are not independent processes, $\mathrm{Im}(N)$ also affects scattering but in a nonlinear way: if a scatterer is much smaller than the wavelength, scattering is proportional to $|N-1|$, i.e., increasing absorptivity increases also scattering; if a scatterer is much larger than the wavelength, increase in $\mathrm{Im}(N)$ decrease the amount of energy scattered, until at sufficiently large $\mathrm{Im}(N)$'s, it becomes increasingly difficult for light to get inside the scatterer, increasing scattering and decreasing absorption.

In general, $\mathrm{Re}(N)$ is easier to measure than $\mathrm{Im}(N)$ (e.g., Patterson et al., 1977). Especially for $\mathrm{Im}(N)$, the wavelength dependence can be strong, so the values need to be measured with sufficiently fine wavelength resolution. Fortunately for meteorological applications, the refractive indices of water and ice, which are the most common substances in the atmosphere, are well known for a wide wavelength range (e.g. Ray, 1972; Warren, 1984). Aerosol particles are more problematic, but much effort is put to get sufficient information also on their refractive indices (see, e.g., d'Almeida et al., 1991; Hess et al., 1998, and references therein). In many meteorological applications, at least $\mathrm{Re}(N)$ can be considered a known parameter.

The concept of refractive index implicitly includes the assumption that the material is locally homogeneous. How the combined effect of atoms and molecules can be characterized macroscopically by using a single (complex) value is described by the so-called optical theorem (see, e.g., Jackson, 1975). Indeed, even macroscopic inhomogeneity can be characterized using a single (effective) refractive index if the size scale of inhomogeneity is smaller than the wavelength of interest (see, e.g., Sihvola, 1989; Chýlek et al., 2000, and references therein). This is called an effective-medium approximation (EMA).

If a scatterer is in a medium, scattering is characterized by a so-called relative refractive index m, which is the ratio of the refractive indices of the scatterer and the medium. Absorption, on the other hand, depends always on N. Fortunately, for most gases in typical conditions, the real part of refractive index is very close to unity and the imaginary part is very small, so that the relative and the absolute refractive indices of airborne particles are practically the same. It is noted that if a scatterer is located in a strongly absorbing medium, the scattering problem becomes quite complicated.

Geometrical factors of interest are the size parameter, the shape, and the orientation of the particle. The size parameter x is defined

$$x = \frac{2\pi r}{\lambda_0} = k_0 r, \tag{2.2}$$

where r is the radius of the particle, and λ_0 and k_0 are the wavelength and the wavenumber, respectively, in free space. The angular distribution of scattered light depends on the size parameter, while the particle size directly only affects the amount of light scattered. Thus, the size parameter is the essential parameter instead of the size itself. The size parameter is well defined only for spherical scatterers; for nonspherical particles r is usually replaced with a radius of sphere with equivalent volume, equivalent surface area, or equivalent cross-sectional surface area. Typically, equivalent-volume approach works best for $x \ll 1$ and equivalent cross-sectional surface area approach for $x \gg 1$. Indeed, the dependence of scattering on the size parameter is so different for different x that the size parameter space is customarily divided into three domains (or regimes): Rayleigh domain ($x \ll 1$), resonance domain ($x \approx 1$), and ray-optics domain ($x \gg 1$).

Size distributions are generally characterized by the so-called effective radius r_{eff} and the effective variance (of radius) ν_{eff}, defined by Hansen and Travis (1974) as

$$r_{eff} = \frac{1}{\langle S \rangle} \int_{r_{min}}^{r_{max}} \pi r^3 n(r) dr, \tag{2.3}$$

$$\nu_{eff} = \frac{1}{\langle S \rangle r_{eff}^2} \int_{r_{min}}^{r_{max}} (r - r_{eff})^2 \pi r^2 n(r) dr, \tag{2.4}$$

$$\langle S \rangle = \int_{r_{min}}^{r_{max}} \pi r^2 n(r) dr, \tag{2.5}$$

where $\langle S \rangle$ is the ensemble-averaged geometric cross-sectional area and $n(r)$ is the size distribution function. Both these quantities are defined under the assumption that scattering is proportional to the cross-sectional surface area and are strictly valid only for non-absorbing particles with large size parameter. When the assumption is valid, the quantities are often sufficient to characterize scattering by a size distribution, regardless of the actual shape of the distribution (Hansen and Travis, 1974). Unfortunately, these quantities are often used when the assumption does not hold. An effective size parameter x_{eff} can be defined by replacing r with r_{eff} in Eq. (2.2).

The orientation of a particle can be described with the angles of incidence, ϑ_i and φ_i, which are standard spherical coordinates. Similarly, scattered light can be described in terms of scattering angles θ_s and ϕ_s. These angles can be defined, e.g., as shown in Fig. 2.1. Let a unit vector $\hat{\boldsymbol{e}}_i$ define the direction of incident radiation (ϑ_i, φ_i) and $\hat{\boldsymbol{e}}_s$ the direction of scattered radiation (θ_s, ϕ_s). The plane defined by these vectors is called the scattering plane and it is unique except when either exact forward- or backscattering is considered. The scattering angle θ_s is the angle between $\hat{\boldsymbol{e}}_s$ and $\hat{\boldsymbol{e}}_i$ in the scattering plane. The azimuthal scattering angle ϕ_s defines the angle between $\hat{\boldsymbol{e}}_\vartheta$ and the projection of $\hat{\boldsymbol{e}}_s$ in the plane defined by vectors $\hat{\boldsymbol{e}}_\vartheta$ and $\hat{\boldsymbol{e}}_\varphi$, e.g., the rotation of the scattering plane with respect to the reference coordinate system given by $\hat{\boldsymbol{e}}_\vartheta$ and $\hat{\boldsymbol{e}}_\varphi$.

The total amount of scattered light is described with a scattering cross section C_{sca}, which corresponds to the surface area into which energy equal to the scattered energy is incident upon, and can be expressed as a product of the (dimensionless) scattering efficiency and the geometrical cross section of the particle. Similarly, we can define an absorption cross section C_{abs} and an extinction cross section $C_{ext} = C_{sca} + C_{abs}$. The so-called single-scattering albedo is defined as $\varpi = C_{sca}/C_{ext}$ and is often used as a measure for the absorptivity of a scatterer.

As stated, the mathematical presentation of scattering process depends on the presentation of radiation. Generally the most convenient way is to use the

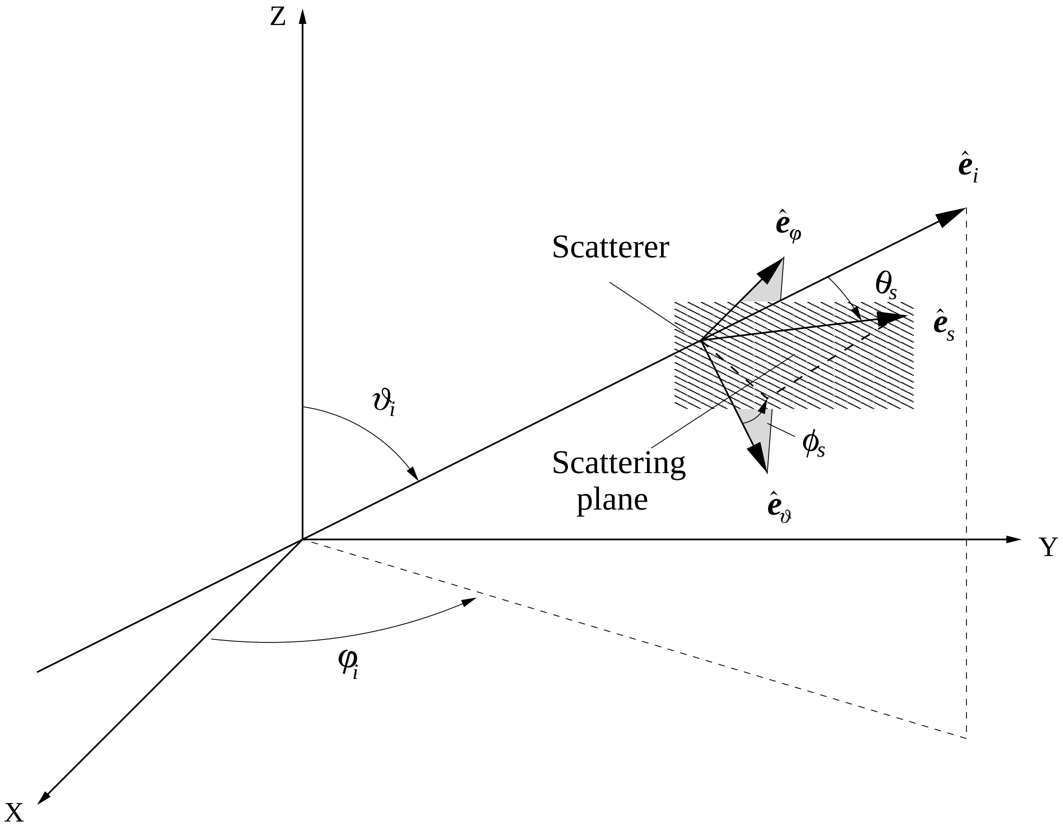

Figure 2.1. Definition of the scattering angles θ_s and ϕ_s, the angles of incidence ϑ_i and φ_i, and the scattering plane. The unit vectors $\hat{\boldsymbol{e}}_i$ and $\hat{\boldsymbol{e}}_s$ define the directions of the incident and scattered radiation, respectively. The components of the incident radiation with respect to the spherical reference coordinate system are given by the unit vectors $\hat{\boldsymbol{e}}_\vartheta$ and $\hat{\boldsymbol{e}}_\varphi$.

so-called Stokes parameters, defined by

$$\begin{aligned} I &= \tfrac{k}{2\omega\mu_0}(E_{||}E_{||}^* + E_\perp E_\perp^*) \\ Q &= \tfrac{k}{2\omega\mu_0}(E_{||}E_{||}^* - E_\perp E_\perp^*) \\ U &= \tfrac{k}{2\omega\mu_0}(E_{||}E_\perp^* + E_\perp E_{||}^*) \\ V &= \tfrac{k}{2\omega\mu_0}i(E_{||}E_\perp^* - E_\perp E_{||}^*), \end{aligned} \tag{2.6}$$

where ω is the angular frequency, μ_0 the permeability of free space, * the complex conjugate, $||$ and $\perp$ stand for the parallel and the perpendicular component in a reference plane, and E is the amplitude of the electric field. The Stokes parameter I is the intensity of radiation, Q/I and U/I define together the degree and the direction of linear polarization, and V/I is the circular polarization. The degree of polarization for arbitrarily polarized incident radiation is given by $\sqrt{Q^2+U^2+V^2}/I$, and for linear polarization specifically it is $\sqrt{Q^2+U^2}/I$. In this thesis, the Stokes parameter formalism is chosen because Stokes parameters can be easily measured (their time averages are meaningful and useful), and they are additive: this is useful when considering ensembles of particles or orientations or, e.g., the net effect of different optical elements (Bohren and Huffman, 1983).

By applying the Stokes parameters, we can write the relation of the incident and the scattered light by

$$\begin{pmatrix} I_s \\ Q_s \\ U_s \\ V_s \end{pmatrix} = \frac{1}{k^2R^2} \begin{bmatrix} S_{11} & S_{12} & S_{13} & S_{14} \\ S_{21} & S_{22} & S_{23} & S_{24} \\ S_{31} & S_{32} & S_{33} & S_{34} \\ S_{41} & S_{42} & S_{43} & S_{44} \end{bmatrix} \begin{pmatrix} I_i \\ Q_i \\ U_i \\ V_i \end{pmatrix}, \tag{2.7}$$

where i and s stand for the incident and the scattered light, respectively, R is the distance from the scatterer, and S_{pq} are the elements of the so-called scattering matrix.

The definition of the scattering matrix is by no means unique. Indeed, there are several differently defined scattering matrices which vary in terms of which quantities are included in the matrix. The scattering matrix $\mathbf{S}$ is convenient if there is need to add several scattering matrices together, e.g., when computing orientation or ensemble-averaged scattering properties, for $\mathbf{S}$'s are additive. Another scattering matrix is the so-called scattering phase matrix $\mathbf{P}$ which is normalized so that the P_{11} element, the so-called phase function, can be used as a probability density function. This is useful, e.g., in radiative transfer applications. The relation between $\mathbf{S}$ and $\mathbf{P}$ is written

$$\mathbf{P} = \frac{4\pi}{k^2 C_{sca}} \mathbf{S}, \tag{2.8}$$

$$C_{sca} = \frac{1}{k^2} \int_{(4\pi)} S_{11} \, d\Omega, \tag{2.9}$$

where Ω is a solid angle.

The phase function P_{11} describes the angular dependence of scattered energy for an unpolarized incident light. Its general shape is often characterized by the so-called asymmetry parameter

$$g = \int_{(4\pi)} \frac{P_{11}}{4\pi} \cos\theta_s \, d\Omega, \tag{2.10}$$

which varies in a range $g \in [-1, 1]$. The degree of linear polarization for unpolarized incident radiation is given as $-P_{12}/P_{11}$ (note the sign difference between this and the case with arbitrarily polarized incident radiation defined earlier: this is a historical convention). The so-called depolarization ratio, describing the decrease of the degree of linear polarization in a scattering process, is defined as $1 - P_{22}/P_{11}$ and varies from 0 (no depolarization) to 1 (full depolarization).

The scattering process is fully known when the scattering matrix $\mathbf{S}$ and the absorption cross section C_{abs} are known. In principle, these quantities include all the information about the scattering process, e.g., all the appropriate properties of the scatterer and the geometry. In a direct scattering problem, the scattering matrix and the absorption cross section are computed theoretically for given particle, radiation, and geometrical characteristics; even this is a difficult task, as there is no general solution for the problem. In the inverse problem, the scattering matrix or part of it is known and the problem is to solve the properties of the scatterer from it; inverse scattering problems can, in practice, only be solved numerically and require also solving the direct problem.

2.2 Geometrical aspects

The geometrical factors relevant for scattering are of special importance in this thesis, so they are discussed more thoroughly here. Basically, there are two kinds of geometrical factors: those that define the scatterer and those that define the scattering process. The most important geometrical characteristics of the scatterer are the shape, the size, and the internal structure, whereas the geometry of the scattering process is described by the angles of incidence and scattering.

How scattering depends qualitatively on particle size, shape, and scattering angle is explained schematically in Chapter 1 of Bohren and Huffman (1983). By subdividing the particle into small regions that scatter light independently of other regions, it is easy to understand how increasing size parameter leads to increasing phase differences between different regions and thus constructive and destructive interference at different scattering angles. In the forward scattering direction, there is mostly constructive interference, as all parts of a scatterer within a given ray path scatter in the same phase there. The larger the size parameter, the closer to the forward direction the constructive interference is limited, and accordingly, the narrower the forward scattering peak. Outside the forward direction, angles of constructive and destructive interference depend on the size parameter and the particle shape, and accordingly, the interference structure can largely be removed by introducing a shape or a size distribution. For very small size parameters, there is hardly any interference in any direction, indicating that the size parameter and the shape have only a weak effect on scattering, and that the angle dependence of scattering is weak. These observations are qualitatively true, although it must be emphasized that in reality the different regions do not scatter light independently.

These same general features are illustrates in Figure 2.2, where an interference of four independent point scatterers forming a square is shown. It is clearly seen how the angular variation of interference is increased when the distance of the point sources compared to the wavelength increases; at the same time, the zone of constructive interference in the forward scattering direction becomes narrower. It is also clearly shown how the interference field near the sources is complex, whereas in the distance the field assumes spherical form. In light scattering applications,

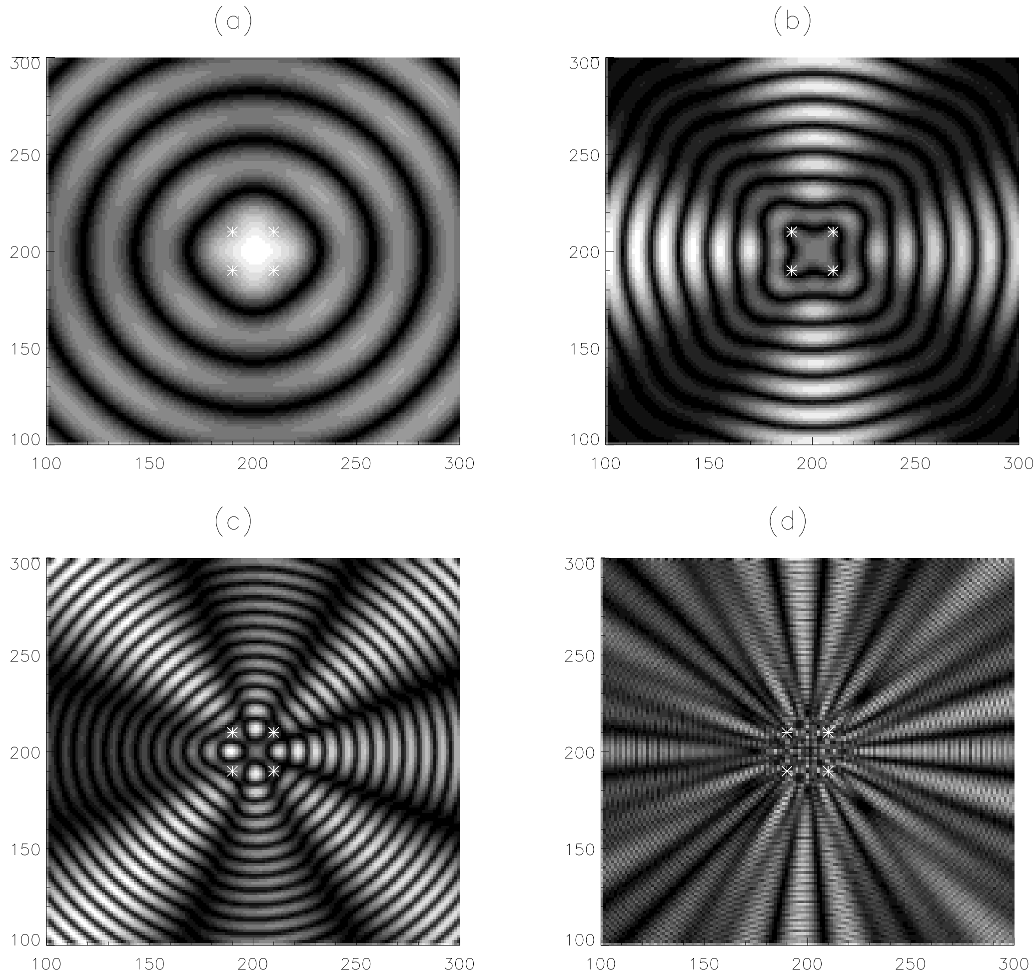

Figure 2.2. Schematic presentation of the interference fields of four independent point scatterers (asterisks) forming a square with a width and height of (a) 0.4λ, (b) 0.75λ, (c) 1.33λ, and (d) 4.0λ. The incident light goes from left to right in each picture. Dark radial segments mark destructive interference and bright radial segments contructive interference. For simplicity, the scattered amplitudes are assumed constant.

the field far away from a scatterer, called the far field, is of main interest.

Modeling of particle size and orientation is straightforward. The modeling of shape, on the other hand, can be quite difficult. Most natural particles, excluding small liquid droplets, are irregularly shaped. For example, natural mineral particles have varying shapes, a surface filled with structures in many scales, they can have internal structures, and they often compose of different minerals, some of which can affect scattering considerably even in small quantities. Similarly, natural ice crystals have seldom simple regular shapes; instead, they often have air bubbles of varying shapes inside them, the prism angles and the width of facets can vary, and there can be surface irregularities due to, e.g., particle collisions and riming (e.g., Yang and Liou, 1998). Not only is it difficult to express an irregular shape simply and exactly, but each particle usually also has its own unique shape. Clearly, as already noted by Bohren and Huffman (1983), when modeling shapes of such particles, a statistical approach is called for. It is also obvious that when different model particles are allowed to have different shapes, one needs to solve ensemble-averaged scattering properties.

Indeed, a statistical shape modeling is one of the key elements of this thesis. Except in Paper III, this is accomplished by applying the Gaussian random sphere geometry (note that Gaussian random spheres are called Gaussian random shapes in Paper I; later, the term Gaussian random sphere has been adapted to describe Gaussian random shapes in a spherical geometry). Similarly, one can define, e.g., Gaussian random cylinders and Gaussian random planes (Muinonen and Saarinen, 2000). Here, this model is described rather generally (for a thorough description, see e.g. Paper I or Muinonen (2000b)). Instead, the focus is in the application of this model to real atmospheric particles.

The Gaussian random sphere model has three properties that have made it a very successful approach in modeling shapes of many natural objects: 1) it is a statistical model, 2) it uses Gaussian statistics, and 3) the radius function is given in an exponential form. A statistical approach is very reasonable when particles with varying shapes need to be modeled. It is not simple to describe accurately even a single nonspherical shape unless it conforms to one of the analytical forms such as an ellipsoid or a Chebyshev shape (e.g., Mishchenko et al., 2000a). Obviously, such simplified nonspherical shapes are exceptions in nature. However, when a whole population is considered instead of a single particle, the situation becomes much simpler. One can then describe the shapes with statistical parameters. With the Gaussian random sphere model, one only needs a mean radius and a covariance function (called autocovariance in Paper I; later, the shorter term has been adapted) of (log)radius. With these parameters, one can control the general type and the degree of nonsphericity in an easy manner, and generate random shapes conforming to the given statistics. However, it means that one cannot control directly the shapes of individual particles, and indeed, statistics of single shapes and the whole population are often clearly different. While this may seem a drawback, it can actually be beneficial: it is much simpler to derive the statistical properties of a population than the exact properties of single particles, and one needs only one set of parameters to characterize the whole population. In addition, if the problem at hand allows, the difference between single-particle and the population statistics can be minimized by a proper choice of covariance function.

The use of Gaussian statistics make the random spheres more relevant for real particles. It follows from the Central Limit Theorem that if particle shapes are a net result of repeated random forcing, the resulting shape is likely to favor Gaussian statistics; in spherical geometry, the lognormal probability density holds for the Central Limit Theorem. The exponential form for the radius vector, on the other hand, guarantees that negative radii are not possible even in the case of strong deformations, so generated particles are realistic. It is also obvious from the Taylor expansion of exponent function that, for small deformations, the direct expansion and the exponential expansions are quite similar: $\exp(y) \approx 1 + y$.

The radius vector for the Gaussian random sphere is defined as

$$\boldsymbol{r}(\vartheta, \varphi) = \frac{a}{\sqrt{1+\sigma^2}} \exp\left[s(\vartheta, \varphi)\right] \hat{\boldsymbol{e}}_r, \tag{2.11}$$

$$s(\vartheta, \varphi) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} s_{lm} \, Y_{lm}(\vartheta, \varphi), \tag{2.12}$$

where ϑ and φ are the spherical coordinates, a the mean radius, σ the relative standard deviation of the radius vector, s the so-called logradius, and $\hat{\boldsymbol{e}}_r$ a unit vector pointing outward in the direction (ϑ, φ). The logradius s, which defines the shape of individual particles (single realizations of the Gaussian random sphere), is given as a real-valued series expansion of spherical harmonics Y_{lm} with degree l and order m. The (complex) weights s_{lm} depend statistically on the covariance function of logradius (Σ_s) which is related to the covariance function of radius ($a^2\Sigma_r$) as $\Sigma_r = \exp(\Sigma_s) - 1$.

Single realizations of the Gaussian random sphere are generated by randomizing the weights s_{lm} as explained, e.g., in Paper IV and Muinonen (1998). Note that two seemingly different forms are given for the spherical harmonics expansion of s in the papers included in this thesis. In Papers I and II, it is given in the form

$$s(\vartheta, \varphi) = \sum_{l=0}^{\infty} \sum_{m=0}^{l} P_l^m(\cos\vartheta)(a_{lm}\cos m\varphi + b_{lm}\sin m\varphi), \tag{2.13}$$

where P_l^m's are the associated Legendre functions and a_{lm}'s and b_{lm}'s are the weighting coefficients. In Papers IV and V, on the other hand, it is given as Eq. (2.12). These two forms are identical, as long as the weighting coefficients s_{lm} are defined properly. The Eq. (2.13) is automatically real-valued as long as a_{lm}'s and b_{lm}'s are real, while Eq. (2.12) is real-valued only if $s_{l,-m} = (-1)^m s_{lm}^*$.

Although the papers included in this thesis consider particles with different shapes, they all incorporate the Gaussian random sphere model, with the exception of Paper III in which a different statistical shape model is used. As the difference in the shapes arises from different covariance functions used, it is reasonable to discuss the concept and the use of the covariance function in detail.

A covariance function is a product of a correlation function and a variance. In general, the correlation function describes the correlation of a quantity or quantities as a function of its parameter and varies in a range of $[-1, 1]$. In the case of the Gaussian random sphere, the quantity is the (log)radius, and the parameter is the angle γ, so the correlation function describes the correlation of (log)radii separated by the angle γ computed over all realizations. If the correlation function is constant (unity), the resulting generated shapes are spheres (with lognormal size distribution, unless $\sigma = 0$). The faster the correlation drops from unity with increasing angle, the smaller-scale features can be in the shape (see Fig. 2.3). Naturally, at 0° the correlation is always unity. The variance, on the other hand, defines the amplitude of the variation. Obviously, the larger the variance, the more the radii vary. For Σ_r, the variance is given as $a^2\sigma^2$, whereas for Σ_s it is $\ln(\sigma^2 + 1)$.

In the Gaussian random sphere model, the correlation function C is convenient to express as a series expansion of Legendre polynomials P_l, e.g.,

$$\Sigma_s(\gamma) = \ln(\sigma^2 + 1)\ C_s(\gamma), \tag{2.14}$$

$$C_s(\gamma) = \sum_{l=0}^{\infty} c_l\ P_l(\cos\gamma), \tag{2.15}$$

where c_l are the weights of Legendre polynomials, as there is a close connection between the Legendre expansion of the correlation function and the spherical harmonics expansion of the logradius: each s_{lm} depends only on the c_l of the corresponding degree (for details, see e.g. the Eqs. (3), (5), and (6) of Paper IV). For the Legendre expansion to be valid, it is further required that

$$\sum_{l=0}^{l_{max}} c_l \equiv 1, \tag{2.16}$$

where l_{max} indicates the maximum degree included in the expansion. All Legendre polynomials are suitable as a correlation function, as they equal to unity at $\gamma = 0°$ and vary in a range $[-1, 1]$ (see, e.g., Arfken, 1985). Indeed, a single-Legendre-polynomial correlation function results in generated shapes consisting only of spherical harmonics of the corresponding degree. The degree of spherical harmonics, on the other hand, directly indicates the number of minima and maxima in the spherical harmonics function, and accordingly, the hills and valleys the appropriate spherical harmonics causes in a shape. Thus, if a shape with small-scale variability in the radius function is desired, high-degree spherical harmonics are needed, and vice versa.

For the spherical harmonics, and correspondingly for the Legendre expansion of the logradius, two lowest degrees are special: the degree $l = 0$ corresponds to a change in the mean radius a, i.e. it does not affect shape; similarly the degree $l = 1$ is almost (almost, because the spherical harmonics expansion is in the exponent) a pure translation, i.e. it moves the shape with respect of the origin without altering the shape itself significantly, unless σ is very large. All higher-degree terms contribute mainly to the shape.

The correlation functions used in the papers included in this thesis will now be reviewed. In Paper I, the so-called modified Gaussian correlation function of logradius (called spherical 'Gaussian' autocorrelation function in Paper I) was applied. This correlation function has the form

$$C_s(\gamma) = \exp\left(-\frac{2}{\ell^2}\sin^2\frac{1}{2}\gamma\right) \tag{2.17}$$

$$\Gamma = 2\arcsin\left(\frac{1}{2}\ell\right), \tag{2.18}$$

where ℓ is the so-called correlation length and Γ is the correlation angle. Thus, the correlation function (of logradius) is controlled by a single parameter ℓ, and the covariance function (and thus the shapes of generated realizations) by ℓ and the variance σ^2. The value of ℓ is conveniently given by using an auxiliary parameter Γ, which represents the angle in which the correlation drops to $1/\sqrt{e}$. Due to its intuitive parameters, the dependence of the shapes of generated realizations on the correlation function can be conveniently demonstrated using the modified Gaussian correlation function; this is shown in Fig. 2.3. Note how the radius varies less within a single realization as Γ increases, although σ is not changed.

The drawback of the modified Gaussian correlation function is that the spherical harmonics degrees $l = 0$ and $l = 1$ are included. They are not important for the shape, and in some cases they are unrealistic (e.g., such oscillation modes would be unphysical). The degree $l = 0$ is especially problematic if a size variation cannot be allowed. Indeed, in Paper IV, in which the modified Gaussian correlation function is also used, the c_0 term has been eliminated from the correlation function to suppress the size variation. The main effect of this suppression is that the possible values for Γ are now restricted, because increasing Γ beyond 60° would only increase c_0 and decrease all other c_l's: if c_0 term is set to zero, such a situation would not satisfy Eq. (2.16). It is emphasized that the removal of the $l = 0$ term does not eliminate the volume variation completely, as the particle volume depends slightly also on higher-degree variations.

In Paper II, single Legendre polynomials with degrees from $l = 2$ to 5 were used as a correlation function of logradius, i.e., one of the c_l is unity and the others are zero. It is emphasized that in Paper II the Gaussian random sphere geometry is used only for the oscillation part of the shape, embedded on a size-dependent equilibrium shape.

Paper V introduced yet another correlation function, this time one that is based on measurements. As explained in the paper, the analysis of Saharan particle silhouettes show that for $l \geq 2$, $c_l \propto l^{-\nu}$, where $\nu \approx 4$. The weights for degrees $l = 0$ and 1 have been set to zero (the value of c_0 does not affect the particle shapes at all; c_1 affects slightly, but its derivation would require quite sophisticated analysis method, such as that explained in Lamberg et al. (2001)). This kind of correlation function, called the power law correlation function, appears quite universal for irregular shapes (Muinonen, 2002b): the same $c_l \propto l^{-4}$ power law holds surprisingly well also for asteroid shapes and the shape of (the gravita-

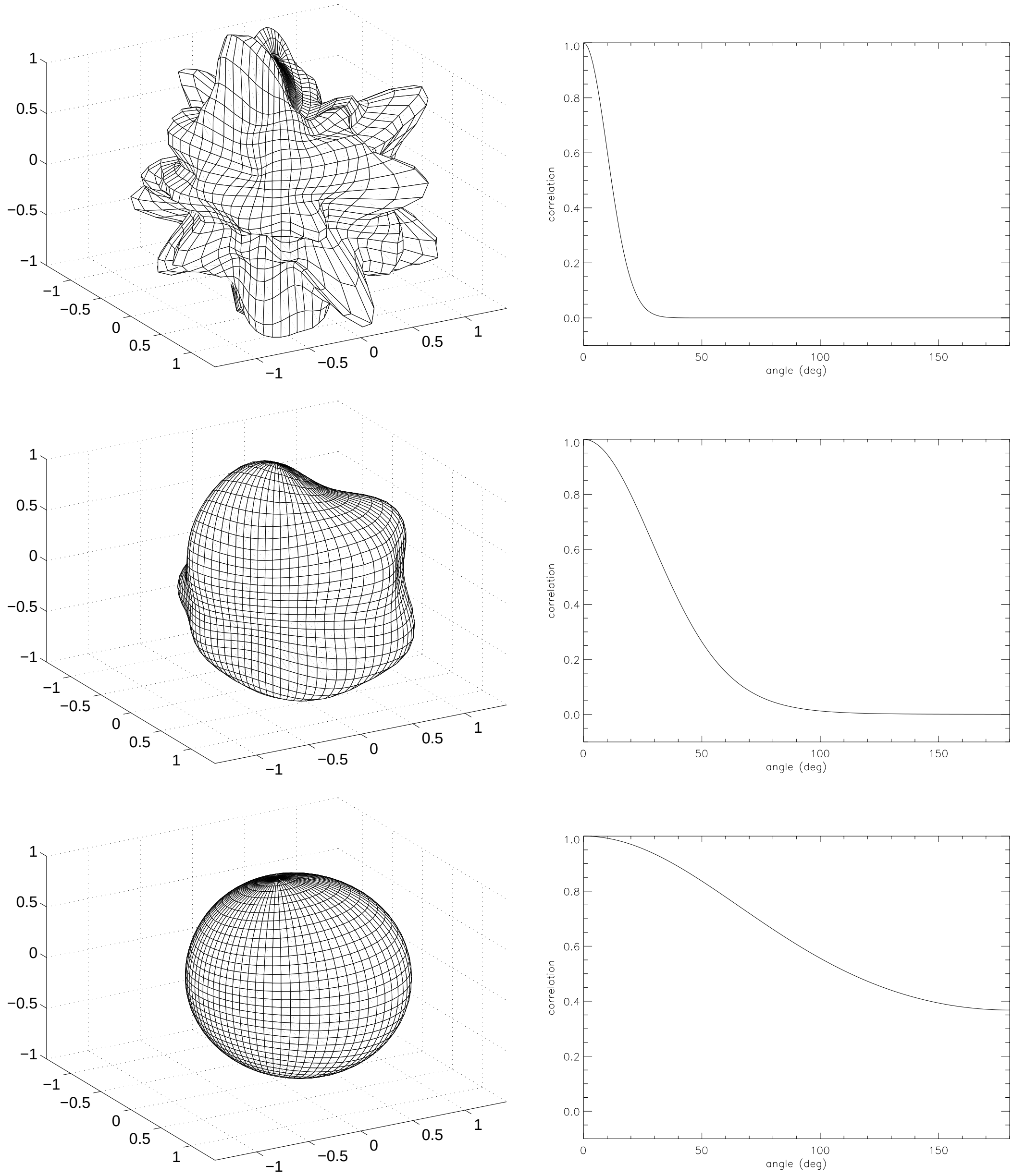

Figure 2.3. Example realizations of Gaussian spheres and the corresponding correlation functions of logradius, generated using the modified Gaussian correlation function with $\sigma = 0.2$ and varying Γ. On the top panel, $\Gamma = 10°$ has been used, while the middle and the bottom panels correspond to $\Gamma = 30°$ and $90°$, respectively.

tional field of) Earth, for example (Kaula, 1968; Muinonen and Lagerros, 1998). Naturally, the relative variance of the radius is very small for Earth compared to asteroids or dust particles. It is quite possible that the power law correlation function with $c_l \propto l^{-4}$ can be used to generate realistic overall shapes for typical natural mineral particles within sufficient accuracy for light scattering problems.

Figure 2.4 illustrates the performance of the Gaussian random sphere model in describing natural mineral dust particles. Although the Saharan particle in the photograph is clearly larger ($a \approx 500\ \mu$m) than those considered in Paper V, it looks quite similar. Unfortunately, the CCD images used for the shape analysis presented in Paper V have been erased years ago, so they cannot be used here. The realization of the Gaussian random sphere shown on the right is generated using the power law correlation function with $\nu = 3.5$ and $l_{max} = 50$, so ν is slightly smaller than the value derived from the shape analysis. A smaller value has been used to increase the small-scale irregularity so that it would more resemble that of real particles. This has the side effect of making the particle shape a fractal, so that the Legendre expansion of the correlation function is no longer convergent. There are alternative ways to increase the small-scale surface irregularity so that the low-degree terms of the correlation function, for which the shape analysis was limited to, are in agreement with the shape analysis, but this issue is not considered further here.

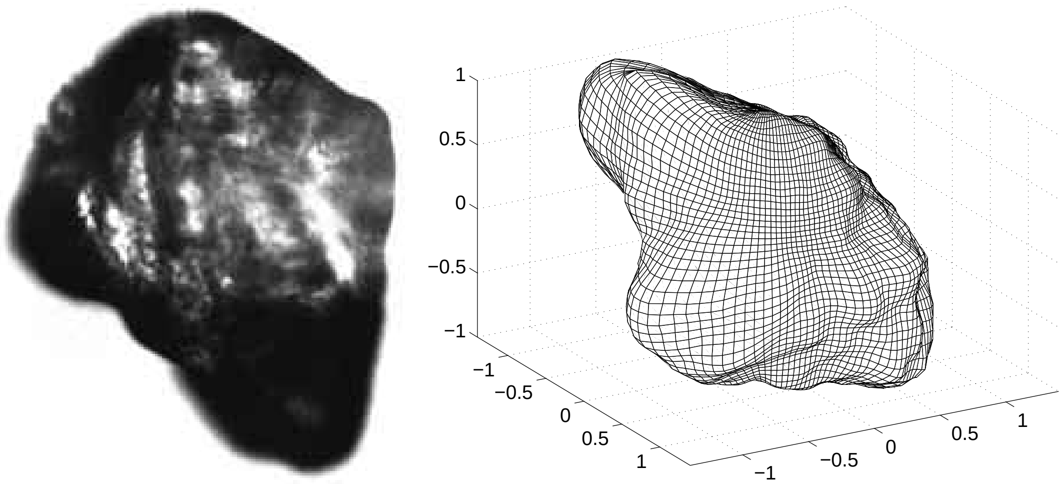

Figure 2.4. A photograph of a Saharan sand grain with $a \approx 500\ \mu$m collected in Libya (left), compared with a realization of a Gaussian random sphere generated with parameters $\sigma = 0.2$ and $l_{max} = 50$ and a power law correlation function with $\nu = 3.5$ (right). The particle looks very dark because it is photographed against an illuminated class plate. (Photograph by Sanna Kaasalainen, Jukka Piironen, and Martti Lehtinen).

Particle orientation is also an important factor when light scattering by non-spherical particles is considered. If scattering by a single particle is considered, the orientation must be fully accounted for. Scattering depends on the angles of incidence (ϑ_i, φ_i) and the resulting scattering pattern is a function of both θ_s and ϕ_s. Thus, in effect, scattering matrices are functions of four angles. On the other hand, if scattering by a population of particles is considered, i.e., ensemble-averaged scattering properties are looked for, there are two different cases to be discussed. If particles are randomly oriented, then the properties of the population are not changed by rotations and scattering matrix for the ensemble is only a function of θ_s. If particles are oriented, however, the situation resembles that of a single particle. If particles can be considered randomly oriented with respect to the azimuthal incident angle φ_i (e.g., horizontally oriented ice crystals), the resulting scattering matrix is a function of angles ϑ_i, θ_s, and ϕ_s. If particles are also azimuthally oriented, then all four angles are needed. So, in this case, a non-random orientation increases the number of variables in the ensemble-averaged scattering matrix from one to three or four. Each additional free parameter doubles the number of necessary computations in a systematic study, so additional variables generally make such studies very laborious. In addition, accounting for non-random orientation complicates statistical shape modeling; this could be done, e.g., by computing the principal moments of inertia for each realization (see Muinonen, 1998) and orienting the particles with respect to these. Obviously, both the single-scattering computations and applying their results is more complicated in case of oriented scatterers.

In addition, the orientation affects the general symmetry properties of the scattering matrix. For a collection of oriented particles, all 16 scattering matrix elements can be non-zero and independent. For a collection of randomly oriented particles, however, there are only 8 non-zero and 6 independent scattering matrix elements (unless particles are composed of optically active material; such considerations are left outside of this thesis).

The effect of orientation on single-scattering studies can be seen in practice by comparing results in Paper I with those in Papers II and III. In Paper I, ensembles of randomly oriented particles are considered, and accordingly, there is only one angle dependence to be considered. In Papers II and III, particles are oriented and scattering depends on the angles ϑ_i, θ_s and ϕ_s. This means that scattering needs to be solved for different angles of incidence, increasing largely the amount of data to be studied. In addition, the results of Papers II and III are difficult to illustrate; curves are easier than full 4π plots to interpret accurately. Finally, in Papers II and III, all 16 scattering matrix elements are non-zero and need (in principle) to be considered, as opposed to 6 independent non-zero elements in Paper I.

2.3 Single-scattering models

In principle, the interaction between electromagnetic fields and matter is fully described by Maxwell's equations, material-dependent constitutive relations, and boundary conditions on the particle surface (e.g., Bohren and Huffman, 1983). As noted in Chapter 1, the resulting set of equations does not have a general analytical solution. The equations can be solved numerically, but this is practical only for small size parameters. As a consequence, there exist a multitude of methods which are typically usable only in a narrow range of the parameter space. So, knowledge of different methods and their regime of validity, as well as the skill of choosing the best method for the problem are important parts of the art of light scattering research. The most important parameters to be considered are the size parameter, the particle shape, and the complex refractive index.

There are some cases in which the resulting set of equations can be analytically and exactly solved. The best-known such solution is the Lorenz-Mie theory (often called the Mie theory) for homogeneous, isotropic spheres. The Lorenz-Mie solution is relatively simple, generally fast to compute, and can be applied to spheres with an arbitrary size parameter and refractive index. These properties have made the Lorenz-Mie theory very popular. Indeed, it is widely used in many fields of science, often for scatterers that are neither homogeneous nor spheres. Other analytical and exact solutions include layered spheres, circular and elliptical infinite cylinders, spheroids, and some other simple particles with a rotational symmetry. The common element for these solutions is that they assume particle geometries which are quite idealistic compared with typical natural particles.

There are also a number of different numerical methods, e.g., different surface- and volume-integral methods. Some volume-integral methods are compared in Peltoniemi et al. (1998) using Gaussian random spheres. While numerical methods are usually much slower than analytical solutions, and often also approximations, they extend largely the parameter space for which a solution can be looked for. For a recent review of different single-scattering methods, see Mishchenko et al. (2000b).

Unfortunately, there are still large gaps in the parameter space in which only inaccurate solutions are available. In general, when solving a scattering problem, one has to choose which is more important: an accurate solution for the "wrong" problem, or an inaccurate solution for the "right" problem. Often, one has to make this decision without being able to test the different options against the accurate solution. Fortunately, an increase in computational power and advances in computational algorithms are closing these gaps promisingly.

As to how different parameters affect the availability of an accurate solution, some general rules can be given. For the refractive index, it is typically the better, the closer the value of m is unity. For example, the small-particle approximations (Rayleigh approximation) apply to larger size parameters if m is close to unity (e.g., Rayleigh-Gans approximation), and volume-integral methods do not require as large a number of volume elements. On the other hand, the large-particle approximations (ray optics) work better if m is larger, and especially if $\mathrm{Im}(m)$ is large, the scattering problem can be much simpler. If the medium in which the scatterer is embedded in is (strongly) absorbing, on the other hand, things become very complicated. Such cases are not considered in this thesis. For the size parameter, the most difficult values are around 10–100, for which accurate solutions are practically available only for simple shapes with exact solution. For values smaller than 10, there are many numerical methods that can handle almost arbitrary particles, but these methods tend to become exceedingly slow for larger size parameters. For size parameters larger than 100, the ray-optics approximation is usually sufficiently accurate. It is emphasized that these limiting values are only rough approximations and they depend on other parameters, e.g., refractive index, as well as the available computational resources. For the shape, symmetric and other simple shapes are easier to handle than irregular and more complex shapes, but they also scatter differently.

The lack of a general solution is especially problematic if a scattering problem includes a wide parameter range to be covered, because in practice this means that different methods need to be applied to different parameter values. While this is laborious, it can also introduce additional problems because most of the methods are not exact. As a consequence, solutions obtained by different methods do not necessarily agree well with each other. These sorts of problems are typical in spectroscopy and in problems involving an integration over a size distribution. It is not surprising that methods which can handle a wide parameter space, e.g. the Lorenz-Mie theory, are popular in such applications even if the method used would not be sufficiently accurate.

Only a few different methods have been used in this thesis. These include a ray-optics approximation (ROA), a second-order perturbation series approximation, the Lorenz-Mie theory, and the discrete dipole approximation (DDA). The latter two are used mainly as supplements and for providing comparison data for testing. ROA is used most: it is used in all papers except in Paper IV.

ROA is based on the fact that incident plane waves of infinite extent can be thought to consist of individual and independent rays of light (van de Hulst, 1981). If a scatterer is large enough compared to the wavelength, the interaction of light rays can be solved independently and assuming that each ray interacts with the particle only locally.

There are two distinct ways in which the rays can interact with the scatterer: (1) rays are reflected or refracted at the boundaries where the refractive index is changed, and absorbed inside the particle if the particle is composed of an absorbing material; (2) alternatively, rays are diffracted if they "miss" the particle close enough. The former, called geometric optics, is customarily solved with Monte Carlo ray tracing, while the latter, called diffraction, can be solved by a numerical integration. Geometric optics and diffraction are different both in the appearance and in the dependences. Diffracted rays are confined closely to the forward scattering direction, geometric optics contribute significantly to all directions. In the first place, diffraction depends on the particle size parameter and shape, but not on the material or internal structure of the particle. For geometric optics both the material and the internal structure are important.

In geometric optics, the phase of light rays is not considered i.e. there is no interference. Further, it is assumed that the particle surface can be considered locally a plane for each incident ray. Both the exclusion of phase effects and the assumption of locally planar surface account for the restriction of ray optics to large size parameters. How large the size parameter should be is not well established and it depends on both the accuracy desired and the characteristics of the scatterer. For the most symmetric shape, a sphere, for which phase effects are most important, the size parameter required may be as high as several hundreds (Hansen and Travis, 1974). For particles with less symmetry, such as cylinders, ray optics has been proven to work for size parameters somewhat below a hundred (Mishchenko and Macke, 1999). For irregular shapes, especially when ensemble-averaged scattering properties are considered, the lower limit is expected to be even below that, but the lack of a fast and accurate solution for irregular particles in this size-parameter range makes it very difficult to check this for sure. Also, ray optics performs better with small size parameters if particles are absorbing. This is because, in such a case, scattering is dominated by the external reflection which involves only one surface interaction as opposed to refracted rays. Indeed, in Paper V ray optics has been used for a size parameter as low as 28 with irregular, absorbing particles.

The accuracy of ROA depends also on the scattering angle. Phase effects are most important in the forward and backscattering directions. In the forward direction, scattering is strongly dominated by diffraction which takes phase effects into account and increases the accuracy of the solution as long as the diffraction part is solved accurately. In the backscattering direction, scattering results from geometric optics, which excludes phase effects, and, accordingly, the accuracy of ray optics is poorer in the backscattering direction (see, e.g., Muinonen, 1990). In side-scattering angles scattering results also from the geometric optics part, but there the phase effects are of clearly less importance.

Different versions of the geometric-optics model have been used in the papers involved. In Paper I, the model incorporates randomly oriented particles. In Papers II and III, the model is modified to handle fixed orientation, and in Paper III the model also incorporates a size distribution. Due to different particle geometries, the geometry routines are also different in these models. The model used in Paper V is clearly different: it is a discretized version, i.e., the particles are represented as a wire frame of triangles instead of a continuous function; for details, see Paper V and Muinonen (2000a). Also, the model is modified to account for both the small-scale irregularity of the particle surface and the particle inhomogeneity by using *ad hoc* simple Lambertian schemes. Again, size distributions and a new kind of particle geometry are incorporated in the model.

With the surface Lambertian scheme, rays crossing the surface undergo either a Fresnelian or a Lambertian reflection/refraction, a probability depending on the surface fraction of Lambertian surface elements. Similarly, with the internal Lambertian scheme, internal rays can hit randomly oriented Lambertian screens and undergo a Lambertian reflection/refraction, the probability depending on the mean free path inside a particle. The Lambertian elements and screens also have given plane albedos that are additional free parameters in the model. The introduction of Lambertian schemes is such a fundamental change that it is actually a matter of opinion if the model can still be called a geometric-optics model.

The diffraction parts have also been handled differently in different papers. In Papers I and V, the diffraction part is solved by a numerical integration in the Kirchhoff approximation (Jackson, 1975; Muinonen et al., 1989), assuming that particles are spheres with equivalent cross-sectional surface area. For most practical applications, such an assumption provides sufficiently accurate solution of diffraction. Indeed, if the size parameter is sufficiently large, the forward diffraction peak can be approximated with a delta spike; this approach was taken in Papers II and III.

In Paper IV, a second-order perturbation series approximation was used. This method is an analytical approximation, resembling the Lorenz-Mie theory in a sense that the particle nonsphericity is considered a minor perturbation in the spherical shape, i.e., the shape is given as $r(\theta, \phi) = a[1 + f(\theta, \phi)]$, where f is a deformation function. The effect of deformation on scattering is taken into account by an additional series expansion that modifies the Lorenz-Mie solution. Typically, a perturbation series approach is applicable only to very slightly deformed spheres, but if a statistical approach is taken, i.e., solving ensemble-averaged scattering properties for a statistically given deformation, the perturbation series behaves better and can be analytically solved for, at least, second order (Schiffer, 1989, 1990). In Paper IV, a statistical second-order perturbation series approximation was applied. As a result, rather large deformations could be considered, but only ensemble-averaged scattering properties could be obtained. This is not necessarily a drawback, as the ensemble-averaged scattering properties are what usually are needed, and they are laborious to compute by averaging scattering simulations for single particles in single orientations. Because the Gaussian random sphere model was used for deformation, implying statistically isotropic deformation, the ensemble-averaged properties implicitly include a random orientation of shapes. Finally, it is required that the size parameter is rather small, not much larger than unity at most. For more nonspherical shapes or larger size parameters, a higher-order perturbation series would be needed. Currently, no such model is available.

There are also other methods that might have been used in some of the work involved. In Papers I, II, and III, ray optics was the only practical option. In Papers IV and V, however, other methods would have been, in principle, possible. In Paper IV, the ice particles could have been assumed spheroidal or simple Chebyshev particles and apply the so-called T-matrix method (also called the extended boundary condition method EBCM; see, e.g. Waterman (1971); Barber and Yeh (1975); Mishchenko and Travis (1998)). However, most implementations of the T-matrix method are restricted to rotationally symmetric particles and thus would have limited the possibilities to study the shape dependence of scattering more than the use of perturbation series method did. In addition, the finding that the depolarization depends mainly on the elongation of the shape at these size parameters would have stayed uncovered. Alternatively, we could also have used volume-integral methods such as a discrete dipole approximation (DDA) (e.g. Lumme and Rahola, 1994; Draine, 2000), finite difference time domain method (FDTD) (e.g. Yang and Liou, 2000), or jscat (Peltoniemi, 1996). These methods could handle even more complex shapes than the perturbation series method can, but they all are extremely laborious to compute compared with the perturbation series method, especially because of the very high refractive index of liquid water at microwave wavelengths which forces one to use a very large number of volume elements to get accurate results. The speed of the DDA, for example, was barely sufficient to compute a single set of comparison data that was used in testing the accuracy of the second-order perturbation series approximation. In Paper V, on the other hand, ray optics was the only practical method for the large-particle part, but the small-particle part could have been computed, e.g., with the T-matrix method or one of the volume-integral methods mentioned above. In addition, the second-order perturbation series approximation could have been used. With the exception of the T-matrix method, however, these methods could not have been used for sufficiently large size parameters to reach the lower limit of ray-optics part, and the T-matrix method could not be used in the same particle geometry. In addition, a sophisticated handling of the small-particle part would have made the paper even more lengthy, and a simultaneous fitting of the small- and the large-particle parts to the measurements would have been very complicated. Thus, a sophisticated handling of the small-particle part was left for the future.

3 Main results

In the following, the papers included in this thesis are reviewed. Section 3.1 considers Papers I–III, whereas Paper IV is reviewed in Section 3.2 and Paper V in Section 3.3.

3.1 Scattering of visible light by oscillating raindrops

As explained in Chapter 1, the basis for the study of light scattering by raindrops was largely established in Paper I, in which deformed spheres with large size parameter was studied systematically. The most important findings of Paper I were that (1) increasing nonsphericity (either due to increasing σ or decreasing Γ) results in a decrease of the asymmetry parameter g; (2) further, an increase in σ affected less, the larger the amplitude already is, implying that the scattering matrix might converge to some limiting matrix for "perfectly" irregular shapes; finally (3), the shape mainly affected scattering in the side- and backscattering directions, whereas changing refractive index also affected forward scattering. These results can be considered quite general for irregular scatterers with large size parameter.

Light scattering by raindrops was studied in Papers II and III. The raindrop shape was modeled as a product of an equilibrium base shape and an oscillation part. The equilibrium shape was given as a size-dependent cosine series expansion and was practically the same in both papers. The oscillation part was, however, different. In Paper II, the oscillation part was modeled as a Gaussian random sphere with the mean radius equal to unity, a correlation function of logradius represented by single Legendre polynomials with degrees $l = 2$ to 5 (oscillation with degrees 0 and 1 are physically unrealistic), and $\sigma = 0.1$. In Paper III, random oscillations were replaced by oscillations of single-mode spherical harmonics with sinusoidal time dependence. The size-dependent amplitudes for different modes were derived from the axis ratio data given in Beard and Kubesh (1991), Kubesh and Beard (1993), and Andsager et al. (1999).

Strictly speaking, neither paper adopts truly realistic particle shapes. As explained in Paper II, a randomly oscillating shape is not physical and cannot be used to model, e.g., evolution of drop shape, but it can be considered quite reasonable as a first approximation for light scattering studies. The oscillation scheme adapted in Paper III is more realistic and more physical, but it is still not exactly physical because the volume and thus the mass of a drop is not exactly constant while the drop oscillates. However, both papers adapt drop shapes that are quite relevant for the studies carried out. For example, the volume of drops could be kept constant by renormalizing the mean radius for each drop, but in the absence of absorption, this would have very little effect on the scattering pattern. The random oscillations, in turn, can be considered to model multiple-mode oscillations. It is not uncommon for raindrops to have multiple-mode oscillations (e.g., compare Fig. 1 of Nelson and Gokhale (1972) with Plate 13 of Pruppacher and Klett (1997)).

In Paper II, scattering properties of the non-oscillating equilibrium shape were studied, and compared with scattering by randomly oscillating raindrops. The study was confined to two distinct sizes. In Paper III, a different shape model was used, and size distributions were introduced. For the size distribution simulations, a size-dependent oscillation scheme was implemented. In addition, the time dependence of scattering by a single oscillating raindrop was briefly studied.

Papers II and III revealed that the most pronounced features in the scattering patterns resulted from the equilibrium shape. In addition, single-mode oscillations introduced some new features which were also different for different modes. The random oscillations, on the other hand, smoothed away features. This is consistent with the results in Paper I. Interestingly, such smoothing was not evident even for a collection of differently oscillating drops, as long as each drop was oscillating in a single mode. In principle, then, the angular dependence of scattering could be used to study raindrop oscillations if each raindrop oscillates only in a single mode. It was also found that the asymmetry parameter g is smaller for nonspherical than for spherical drops except for equilibrium drops in some orientations, and both the random oscillations and the single-mode oscillations decreased g.

One interesting finding was the existence of previously undocumented rainbow type, the so-called 90° rainbow, arising from the nonsphericity of the equilibrium shape. As discussed in van de Hulst (1981), ordinary rainbow phenomena, and indeed, even their explanations (apart from minor details), have been known for almost 200 years. Thus, the 90° rainbow, if it could be seen in nature, might be the first new rainbow phenomenon in a very long time. This phenomenon can only be seen for some angles of incidence, but it is sufficiently intense to be seen also in the orientation-averaged scattering patterns. Interestingly, it can be seen in Fig. 3 of Macke and Großklaus (1998), but it has not been identified as an independent phenomenon, because Macke and Großklaus confined themselves to randomly oriented drops and thus to azimuthally averaged scattering patterns. The 90° rainbow, unfortunately, turned out to be strongly size dependent; this and the tendency of large, sufficiently flattened drops to oscillate make it unlikely to see this phenomenon in nature. On the other hand, instantaneous size distributions are clearly more monodisperse than the averaged distributions, so some rare conditions might allow the phenomenon to be seen.

Different size distributions resulted in a similar kind of smoothing in scattering patterns, indicating that the particle size distribution would be very difficult to retrieve from the angle dependence of scattering. The same result, in the absence of oscillations, was found by Macke and Großklaus (1998). The time dependence of scattering was found to be strong and should be taken into account in measurements. If time-dependent measurements can be carried out with sufficient resolution, this can be used to obtain more information.

Finally, the scattering patterns for spherical and even weakly deformed drops were found to be clearly different; the most obvious effect is the dependence on the azimuthal scattering angle ϕ_s and the increased number of non-zero scattering matrix elements for oriented nonspherical drops. In addition, there is a clear difference in the polarization characteristics of features. This results from the total internal reflection which can take place only in nonspherical shapes.

The evaluation of the performance of different oscillation schemes is limited, because the results cannot be compared with measured scattering matrices and because somewhat different aspects were studied in Papers II and III. In addition, one of the smoothing processes, the raindrop canting (variation of drop orientation), is excluded in Paper III, whereas in the random-oscillation simulations it is mimicked by random oscillations. Nevertheless, it appears that the random oscillation scheme is a good approximation for raindrop oscillations mainly in the case of multiple-mode oscillations. However, usefulness of the random oscillation approach increases when integrated scattering properties are considered: azimuthally averaged scattering matrix elements are more usable than the whole 4π elements, and the asymmetry parameter more usable than either azimuthally averaged or the whole 4π elements.

The results of both Paper II and III clearly show that the assumption of spherical shape for raindrops can result in large errors, and accordingly, raindrop shape effects should also be considered in optical applications. Most importantly, the error caused by assuming spherical drops is systematic in the sense that spherical drops are considerably more forward scattering than nonspherical drops, except in few special cases.

3.2 Microwave backscattering by graupel and hail

Microwave backscattering by ice particles of interest was modeled using the second-order perturbation series approximation. This method can handle rather complex particle shapes, as long as they can be considered nearly spherical, i.e., the deviation from sphericity can be considered a minor perturbation. Thus, rather realistic particle shapes could be used and the shape effects studied in a way different from the previous studies. In addition, this method solves the ensemble-averaged scattering properties analytically; this is a major advantage, as they are what a radar usually measures, and ensemble averaging afterwards is typically a very slow process. The model particle shapes were described using Gaussian random spheres, sufficiently modified to be applicable to the perturbation model. The size variation due to random deformation was minimized by modifying the applied modified Gaussian correlation function properly. This was necessary because scattering cross sections are strong functions of size for size parameters as small as those considered here. The handling of composition and internal structure required additional modeling, as these factors cannot be explicitly taken into account by the perturbation method used. Thus, we applied effective-medium approximations (EMA) to replace inhomogeneous material with an equivalent homogeneous material with an effective (relative) refractive index m_{eff} (see, e.g., Sihvola, 1989; Chýlek et al., 2000). The value of m_{eff} depended on the relative amounts of ice, air, and liquid water, the shape of liquid water inclusions (spherical pockets, layered around air bubbles, or needle-like capillarities), and the mixing assumed for the constituents (controlled by the mixing rule used). Thus, the composition and internal structure were interconnected.

Light scattering simulations were carried out at 5.6 GHz ($\lambda = 5.3$ cm), corresponding to the wavelength of Tuulia, the Doppler radar of the Department of Meteorology (presently: Division of Atmospheric Sciences, Department of Physical Sciences), University of Helsinki. In theory, free parameters in the systematic simulations were a, σ, Γ, and m_{eff}. In practice, m_{eff} was an auxiliary parameter, computed for the given combination of composition, internal structure, and mixing. Particle sizes ranged from $a = 0.5$ to 4 mm, corresponding to size parameters $x = 0.059$–0.47. Shape was varied using the values 20° and 40° for Γ and the values of 0.05 and 0.1 for σ, thus confining to sufficiently small deformations so that the perturbation series approximation would hold. The volume fraction of liquid water varied in a range of 0 to 20 vol% and the volume fraction of air in a range of 0 to 40 vol%. However, when the volume fraction of air was varied, the volume fraction of liquid water was kept in constant 0 vol%, and when the amount of liquid water was varied, the air content was kept constant at 10 vol%.

Unlike in the other papers of this thesis, the angular dependence of scattering was not studied. Instead, having radar applications in mind, the study was restricted to backscattering. The quantities of interest were the co- and depolarized backscattering cross sections $\sigma_{\rm COP}$ and $\sigma_{\rm DEP}$, and the linear depolarization ratio LDR which is simply the ratio $\sigma_{\rm DEP}/\sigma_{\rm COP}$ in the case of a collection of randomly oriented particles and backscattering direction.

When particle shapes were varied in the range given above, it was found that $\sigma_{\rm COP}$ was within about 20% of the Lorenz-Mie solution for the corresponding mean radius and within 10% of the Lorenz-Mie solution for the corresponding equivalent-volume distribution. Backscattering was stronger for deformed shapes, with few exceptions. For deformed shapes, $\sigma_{\rm DEP}$ increased about fourfold when σ was doubled. This holds at least in the range of $\sigma = 0.025$ to 0.1. The dependence on Γ was, on the other hand, more complex and this is one of the key results of this paper. It turned out that particles with the most elongated shapes produced the largest $\sigma_{\rm DEP}$, whereas more complex deformations decreased $\sigma_{\rm DEP}$. Thus, $\sigma_{\rm DEP}$ (and, accordingly, LDR) are most sensitive to the elongation of particle shape, whereas smaller-scale irregularities appear to be of secondary importance. This is surprising, as depolarization is traditionally considered a good measure for nonsphericity. This result illustrates the importance of sophisticated shape modeling even for small size parameters, especially when polarization quantities are studied. It also indicates that simplified shapes such as spheroids or general ellipsoids might be surprisingly usable for modeling microwave backscattering by ice particles with small size parameter. For spheroids, there are very efficient scattering methods available (separation of variables, the T-matrix method), whereas for ellipsoids similar methods are still under development. With small size parameters the Rayleigh-ellipsoid approximation (Battaglia et al., 1999) can be used.

The qualitative effect of refractive index on scattering can be well approximated by the Rayleigh theory: scattering strengthens with increasing $|m-1|$. As liquid water content affects m most, it is also a dominating factor for scattering: Indeed, for σ_{COP}, an introduction of 20 vol% of liquid water into moisture-free particles increased scattering by a factor of 2–4 (depending on other parameters), and for σ_{DEP} by a factor of 4.5–15. The air content and the shape of liquid water inclusions were also important factors, although clearly less important than the amount of liquid water. The mixing assumption typically had rather little effect.

It is emphasized that these results apply only to the parameter values indicated above and should not be extrapolated to, e.g., higher liquid water or air contents, larger size parameters, or more nonspherical shapes. For example, there are often much larger ice particles in the atmosphere than those studied here, and there are radars that use significantly shorter wavelength than that of Tuulia. In such cases, the results given here may not be applicable even qualitatively. In addition, the results do not apply to cases with a pronounced liquid water coating on the particle, such as strongly melted particles. Nevertheless, it is obvious that the shape effects need to be taken properly into account when polarization quantities are considered. For co-polarized backscattering, the shape effects are generally too small to affect measurements significantly: the inherent inaccuracy of radar measurements is too large.

3.3 Scattering of visible light by Saharan mineral particles

In Paper V, continuing the preliminary work presented in Nousiainen et al. (1995), the light scattering properties of large mineral particles were studied by comparing light scattering simulations with a scattering matrix of a sample of Saharan mineral particles measured in laboratory by Volten et al. (2001). In the sample, the particle sizes vary from about 80 nm to 180 μm in radius and the size parameter varies in a range of 1.1 to 2560 at $\lambda = 441.6$ nm. The study, however, was restricted to particles $a > 2$ μm ($x > 28$) as the light scattering method adapted would have not worked for smaller particles, and the restriction to a single method simplified the work significantly. In addition, Volten et al. had already shown that ray optics, which can handle complex shapes, can be made to fit the scattering matrix measured better than any other method in previously published studies. The availability of measurement data allowed for a rough derivation of some physical parameters for Saharan particles as an inverse light scattering problem, but the results have to be considered preliminary: the measurements include a contribution from small particles that was not taken into account in the simulations, so the results are subject to some (unknown) uncertainty.

Unlike raindrops, mineral aerosol particles are clearly absorbing at visible wavelengths. For particles with a large size parameter, this means that scattering is dominated by surface reflection. Because surface reflection from a collection of randomly oriented convex scatterers is independent of particle shape (van de Hulst, 1981), it could be expected that the shape is of less importance than, e.g., in the case of raindrops, although there are concavities in natural mineral particles. Indeed, ray-optics simulations confirmed this: particles with $\mathrm{Im}(m)x \gtrsim 0.7$ scattered light very similarly; this corresponds to particles about 5 μm in radius, if $\mathrm{Im}(m) = 0.01$ derived from d'Almeida et al. (1991) for $\lambda = 441.6$ nm is applied. Actually, Mishchenko et al. (1997) show that the same seems to hold even for particles with a relatively small size parameter (the size parameter corresponding to the effective radius of the size distribution in their T-matrix simulations was $x_{\mathit{eff}} = 8.4$), in which case surface reflection cannot be considered an independent process. This result speaks for the applicability of ray optics for surprisingly small size parameters in situations when scattering is dominated by surface reflection.

In order to take the particle shapes into account properly, a tentative shape analysis was carried out to derive the shape statistics of natural mineral particles. The results of this analysis are interesting in themselves, as they indicate that the shapes of natural mineral particles resemble statistically those of asteroids (Muinonen and Lagerros, 1998) or, indeed, the (gravitational) shape of Earth (Kaula, 1968). It seems that there is something very general in shapes of natural irregular objects.

Simulations with traditional (Fresnelian) ray-optics model showed that realistically shaped particles results in scattering with too strong linear polarization and too weak depolarization compared with measurements, even if $\mathrm{Im}(m)$ was decreased considerably from the literature values. Good fits with measurements required the use of a very spiky shape (see also Volten et al., 2001; Nousiainen and Muinonen, 2002). This indicated that Fresnelian surface reflection does not work well for natural mineral particles, i.e., natural mineral particles reflect light more diffusely. Thus, an *ad hoc* simple Lambertian scheme was introduced to study the importance of small-scale surface irregularity on scattering. Similarly, a Lambertian scheme was devised to mimic internal structure. When slightly inhomogeneous model particles with a partially Lambertian surface were used, realistic shapes could be used to obtain a very good agreement between simulations and measurements, although this required decreasing Im(m) somewhat below the value derived from d'Almeida et al. (1991). If Im(m) was not decreased, equally good fits were obtained, but only when using both Lambertian schemes and spiky shapes. The Lambertian surface elements appeared more important than the internal screens for good fits. This is easy to understand, considering the absorptivity of particles and the resulting dominance of surface reflection on scattering. It is emphasized, however, that internal screens alone also improved fits.

Simulations with a Lorenz-Mie model, on the other hand, showed clearly that the assumption of spherical, homogeneous particles is quite dangerous in the case of natural mineral particles. Even the phase function was in error, and the polarization characteristics were quite different from measurements. It appears that the Lorenz-Mie theory is not sufficiently accurate even for those small particles in the size distribution that could not be modeled using the ray-optics model. The use of Lorenz-Mie theory is especially dangerous with nadir-looking satellite instruments, because they generally measure scattering at angles larger than $\theta_s > 90°$, for which the Lorenz-Mie theory gives scattering that can be an order of magnitude in error.

Radiative transfer simulations were used to illustrate the effect of sophisticated single-scattering modeling by comparing results obtained using simple Lorenz-Mie theory to those obtained by using the ray-optics model. Although the test case is dominated by absorption rather than scattering, and the molecular Rayleigh scattering is very strong at the wavelength used, it was shown that both the particle nonsphericity and the value of Im(m) are important for correct results. Further, it is noted that the values of Im(m) for natural atmospheric mineral particles are often measured by using simplified scattering models (see, e.g., Dubovik et al., 2002). Considering the results given here, it is questionable how accurate values can thus be obtained.

It is obvious that when large objects like rocks are considered, accurate scattering modeling requires a proper handling of the surface texture. For example, a glass object can be turned from transparent to white (diffuse scatterer) by roughening its surface properly. While this does not change the global shape of the object, it has a profound effect on scattering. For example, as shown in Paper II, a spherical water drop with large size parameter has an asymmetry parameter of 0.89, whereas for a corresponding perfect "white particle" it is 0.28 (computed from van de Hulst, 1981). While there are no perfect white particles in nature, this example illustrates well the potential impact of surface texture on scattering. The results of Paper V strongly indicate this applies to surprisingly small particles, perhaps even those somewhat below the ray-optics domain. At the same time, it appears that roughening the surface decreases the importance of the large-scale shape on scattering (Nousiainen and Muinonen, 2002). Due to the uncertainties involved, nothing can be said for sure at this point, but if this indeed is true, then the implications are clear: one must use a method that can handle surface irregularity to get accurate results for scatterers with rough surface and large size parameter. It appears that a simple Lambertian modification can be used to improve the performance of ray optics, but the goodness of this approximation cannot be established at this point. It requires a physically rigorous scattering method for comparison, scattering measurements in which all scatterers are well inside the ray-optics domain, or, in case the measurements include also small particles, a proper handling of the small-particle part. The last option is uncomfortable, as one then has to deal with uncertainties involved in, at least, two different methods, without a possibility to study them independently. If it turns out that a more accurate, or a physically more rigorous replacement for the Lambertian modification is needed, it could be based on an approximate multiple scattering treatment within each particle, accounting also for phase effects (see e.g. Muinonen, 2002a). Obviously, such an approach would be quite time consuming. Alternatively, one might model the reflection using, e.g., an infinite plane surface with small scatterers close to it (Muinonen, 1990; Videen, 1992; Ermutlu et al., 1995). It is also noted that the introduction of a modified Kirchhoff approximation for ROA (Muinonen, 1989) reduces polarization especially for the smallest particles in the ray-optics domain. However, a computationally feasible approach introduced here may be of great value in many practical applications.

It is emphasized that the modified ray-optics approach introduced here is not yet ready for general applications, as measured scattering matrices are needed to set values for the Lambertian parameters. The performance of the modified ray-optics model seems impressive, but more work is needed. In addition, before the method can be used for atmospheric applications, it must be combined with another method to handle the particles too small for ray optics. If a method can be developed that can describe scattering by irregular mineral particles of arbitrary size as accurately as is shown in the best fits of Paper V without a need to use measurements in fixing the free parameters, it will have a profound impact on remote sensing applications dealing with natural mineral particles.

4 Discussion

In this thesis, the importance of sophisticated single-scattering modeling in an example set of atmospheric scattering problems are studied. This is an important topic, as too simplified scattering models are often used in meteorology and in other fields of applied physics.

The results of this thesis are in agreement with previous studies showing that the use of too simple a scattering method can introduce large errors in scattering properties, and that polarimetric quantities respond differently and often more sensibly to the particle shape than the intensity does. Thus, whenever polarimetry is involved, one should pay special attention to scattering modeling. In addition, the results show that sophisticated modeling is important both at small and large size parameters. There is a clear indication that, at least in the case of ensemble-averaged scattering properties of irregular particles, the importance of shape on scattering increases with increasing size parameter, until at sufficiently large values the surface texture becomes the dominating factor, provided that the surface is not smooth and featureless. Apparently, this dominance can be sufficiently strong to make scattering considerably insensitive to the large-scale shape.

The angular dependence of scattering is only considered in the case of large size parameters, but to some extend, the results may apply also to smaller particles. As shown in Papers I–III (and a number of previous studies), nonspherically shaped particles with large size parameter generally have asymmetry parameters smaller than the corresponding spherical particles. Thus, assuming a spherical shape for scatterers will likely cause a systematic error when estimating the flux of scattered energy. This is especially important to take into account in radiative transfer applications. In addition, it appears that the effect of size distributions on scattering is practically purely that of smoothing. This makes it very difficult to retrieve size distributions from the angular dependence of scattering. There are also other apparently systematic effects in scattering as the nonsphericity of particles increases; for details, see Papers I and II. The forward scattering direction appears to be clearly less sensitive to detailed particle properties than other directions. In ray-optics, the forward scattering is dominated by diffraction which, in a first place, does not depend on the particle material or the internal structure. However, it seems that even the geometric optics part is insensitive to particle shape at forward-scattering angles. In general, it seems that the refractive index affects scattering in all angles, but the shape affects mainly side- and backscattering. There is one notable exception to this rule: in Paper III the time-dependence of scattering resulting from the time-dependent shape showed some variation in the scattering pattern in the forward direction. This implies that this rule is more valid for irregular than for regular shapes, and is probably due to the absence of focal effects in ensembles of irregular shapes.

The inhomogeneity of particles is considered in Papers IV and V. In Paper IV, it affects scattering indirectly by modifying the effective refractive index and is thus subject to the assumption that effective-medium approximations can be safely used. In Paper V, the inhomogeneity is modeled using an *ad hoc* simple Lambertian idealization. Thus, neither of these papers directly model inhomogeneity, and neither study systematically the importance of the inhomogeneity for accurate scattering modeling. In Paper IV different assumptions about internal structure and composition lead to quite different scattering. The results in Paper V indicate that the inhomogeneity would be of secondary importance, but in this case scattering is dominated by surface reflection so the result should not be generalized. In addition, the internal inhomogeneity alone can also be used to increase the agreement between measurements and simulations in Paper V. This can be qualitatively seen in Fig. 6 of Paper V: decreasing the free path length inside particles drives the phase-matrix elements closer to the measured values (shown, e.g., in Fig. 3 of Paper V). It is also noted that in the case of absorbing particles the exclusion of internal inhomogeneity can bias the single-scattering albedo, especially for particles with a large size parameter. Inhomogeneities can scatter a part of the radiation that is refracted inside the particle back towards the surface before it is significantly absorbed, resulting in higher single-scattering albedos for internally inhomogeneous particles.

Accurate single-scattering modeling is especially important in inverse problems. First, due to nonlinearities, they can respond to small differences in scattering patterns quite unpredictably. Second, polarization characteristics, which are more sensitive to details, provide additional independent information; this is of special importance in inversion problems. In this thesis, inverse light scattering approaches have not been given much weight; the main focus has been in the direct scattering problem. However, the direct problem has been considered keeping in mind the inversion applications. As light scattering simulations have been compared with real measurements only in Paper V, any inversion would have been irrelevant in other papers. In Paper V, rough inversion is carried out: the imaginary index of particles and the values for the Lambertian parameters are estimated by fitting the simulations to measurements.

The lack of general, exact, and efficient solution to a scattering problem means that there is going to be a continuous need for simplifications in future. Indeed, the very concept of modeling means simplified representation of reality. There are two basic approaches that can be taken: one can either simplify the scattering problem or the method used to solve it. The former includes, e.g., assuming idealized shapes for scatterers, while the latter corresponds to using approximative solutions such as a perturbation series approximation or neglecting some physics, e.g., not accounting for phase effects, or applying empirical scattering laws. When, what, and how much to simplify are key questions, but do not have simple answers. One could say that the art of scattering modeling, like all modeling, is largely an art of proper simplification. Not only is it often difficult to estimate which approach provides the most accurate results for a given scattering problem, but one also usually has quite limited computational resources, and thus, may not be able to use the most accurate approach but rather the most cost-effective one. In addition, if scattering computations are carried out for an external application, the application itself may set some extra constraints. For example, most radiative transfer models cannot handle oriented scatterers. Finally, there is the issue of how much is known about scatterers. The more detailed scattering modeling is carried out, the more information on the scatterers is required, or more assumptions about the details need to be made.

Bearing in mind these facts, some guidelines for the single-scattering modeling are suggested:

- The larger the size parameter, the more important small details are. For example, Paper IV indicates that in the case of small size parameter, depolarized backscattering is well modeled using correctly elongated particles, whereas Paper V indicates that in the case of large size parameter even a correct global shape may not be sufficient but the surface texture needs also be taken into account properly.

- When irregular natural particles are considered, a statistical particle model is strongly suggested. This decreases the amount of information needed for scatterers, makes it easier to control the properties of model particles, and decreases a risk of artificial scattering features such as caustics (e.g. haloes and rainbows). Naturally, the statistical model used should be appropriate for the objects modeled, i.e., Gaussian random spheres should not be used for hexagonal ice crystals.

- Even small deviations from a symmetrical shape can be important, especially in the case of large size parameter. On the other hand, differently irregular shapes often scatter light rather similarly. Thus, in the case of irregular particles, it is often better to use wrongly (but reasonably) irregular shapes than to assume a symmetric shape.

- Integration tends to compensate errors, so integrated quantities are often rather insensitive to simplifications. However, the errors introduced by simplifications in particle shapes are usually systematic. Thus, even if only integrated quantities are needed, sophisticated modeling may be quite important.

- Scattering patterns can be quite dependent on the orientation, but on the other hand, the use of non-random orientation makes single-scattering modeling much more laborious and complicated.

- There are few methods which can apply to wide parameter ranges, such as to a wide wavelength range or a wide size distribution. This can cause additional problems, especially if methods with different kind of assumptions are used.

Considering future research, it is apparent that some projects in this thesis could and should be continued, while others represent more or less completed cases. For example, modeling scattering of visible light by raindrops mainly requires more data on raindrop shape and its variations, whereas there appears to be little need for developing the scattering model further. On the other hand, Paper V appears more like an introduction to a new way of handling scattering by natural mineral particles. Whether or not a ray optics augmented with Lambertian schemes is the best way of doing it (even cost-effectively) remains to be seen, but it is clear that more work is needed especially with small particles. The importance of this problem guarantees that more work will be done. Likewise, the results obtained in Paper IV should be tested also with size distributions.

One intriguing "non-scattering" result given in Paper V is the apparent universality of shape statistics of natural irregular particles. Considering that from a light-scattering point of view the exact shape of irregular particles is not as critical as the irregularity itself, the possibility of generating irregular particles that have sufficiently accurate shape statistics allows one to devise a universal mineral particle shape model to be used in scattering simulations. Indeed, it has already been noticed that many natural particles scatter light similarly, and accordingly, an average mineral particle scattering matrix has been suggested to be used in applications (Volten et al., 2001). Along these lines, it appears plausible to use the power law correlation function used in Paper V as a first approximation when modeling light scattering by irregularly shaped natural particles. This is especially reasonable if particle shapes are unknown and cannot be measured, e.g. in case of extraterrestrial mineral particles.

In order to improve the usefulness and the quality of light scattering models, it is crucial to have more measured light scattering data with many different particle classes and size ranges. Both the model development and estimating the performance of models greatly benefit from available high-quality measurements. Paper V is a good example how measurements, although not ideal for the work at hand, can help a model development. The same also holds the other way, measurements need to be compared with simulations, e.g., to calibrate the equipment and to notice possible problems such as misalignment of optical components. In addition, computer models can be used to map the most useful measurement arrangements and the quantities one should pay special attention to.

References

Andsager, K., K. V. Beard and N. F. Laird, 1999: Laboratory measurements of axis ratios for large raindrops. *J. Atmos. Sci.*, **56**(15), 2673–2683.

Arfken, G., 1985: *Mathematical Methods for Physicists.* Academic Press, San Diego, 3rd edn. 985 pp.

Aydin, K., 2000: Centimeter and millimeter wave scattering from nonspherical hydrometeors. In: *Light Scattering by Nonspherical Particles* (edited by Mishchenko, M. I., J. W. Hovenier and L. D. Travis), chap. 16, pp. 451–479. Academic Press, San Diego.

Barber, P. and C. Yeh, 1975: Scattering of electromagnetic waves by arbitrarily shaped dielectric bodies. *Appl. Opt.*, **14**, 2864–2872.

Battaglia, A., K. Muinonen, T. Nousiainen and J. I. Peltoniemi, 1999: Light scattering by Gaussian particles: Rayleigh-ellipsoid approximation. *J. Quant. Spectrosc. Radiat. Transfer*, **63**, 277–303.

Beard, K. V. and C. Chuang, 1987: A new model for the equilibrium shape of raindrops. *J. Atmos. Sci.*, **44**(11), 1509–1524.

Beard, K. V. and A. R. Jameson, 1983: Raindrop canting. *J. Atmos. Sci.*, **40**, 448–454.

Beard, K. V. and R. J. Kubesh, 1991: Laboratory measurements of small raindrop distortion. Part 2: Oscillation frequencies and modes. *J. Atmos. Sci.*, **48**(20), 2245–2264.

Beard, K. V., H. T. Ochs III and R. J. Kubesh, 1989: Natural oscillations of small raindrops. *Nature*, **342**, 408–410.

Bohren, C. F. and D. R. Huffman, 1983: *Absorption and Scattering of Light by Small Particles.* John Wiley & Sons. 530 pp.

Chýlek, P., B. R. D. Gupta, N. C. Knight and C. A. Knight, 1984: Distribution of water in hailstones. *J. Climate Appl. Meteor.*, **23**(10), 1469–1472.

Chýlek, P., G. Videen, D. J. W. Geldart, J. S. Dobbie and H. C. W. Tso, 2000: Effective medium approximations for heterogeneous particles. In: *Light Scattering by Nonspherical Particles* (edited by Mishchenko, M. I., J. W. Hovenier and L. D. Travis), chap. 9, pp. 273–308. Academic Press, San Diego.

d'Almeida, G. A., P. Koepke and E. P. Shettle, 1991: *Atmospheric Aerosols: Global Climatology and Radiative Characteristics.* Deepak, Hampton, VA. 561 pp.

Draine, B. T., 2000: The discrete dipole approximation for light scattering by irregular targets. In: *Light Scattering by Nonspherical Particles* (edited by Mishchenko, M. I., J. W. Hovenier and L. D. Travis), chap. 5, pp. 131–145. Academic Press, San Diego.

Dubovik, O., B. N. Holben, T. F. Eck, A. Smirnov, Y. J. Kaufman, M. D. King, D. Tanré and I. Slutsker, 2002: Variability of absorption and optical properties of key aerosol types observed in worldwide locations. *J. Atmos. Sci.*, **59**, 590–608.

Ermutlu, M. E., K. Muinonen, K. A. Lumme, I. V. Lindell and A. H. Sihvola, 1995: Scattering by a small object close to an interface. III: Buried object. *J. Opt. Soc. Amer. A*, **12**, 1310–1315.

Goddard, J. W. F., S. M. Cherry and V. N. Bringi, 1982: Comparison of dual-polarized radar measurements of rain with ground-based disdrometer measurements. *J. Appl. Meteor.*, **21**, 252–256.

Haferman, J. L., 2000: Microwave scattering by precipitation. In: *Light Scattering by Nonspherical Particles* (edited by Mishchenko, M. I., J. W. Hovenier and L. D. Travis), chap. 17, pp. 481–524. Academic Press, San Diego.

Hansen, J. E. and L. D. Travis, 1974: Light scattering in planetary atmospheres. *Space Sci. Rev.*, **16**, 527–610.

Hendry, A., G. C. McCormick and B. L. Barge, 1976: The degree of common orientation of hydrometeors observed by polarization diversity radars. *J. Appl. Meteor.*, **15**, 633–640.

Herzegh, P. H. and A. R. Jameson, 1992: Observing precipitation through dual-polarization radar measurements. *Bull. Amer. Meteor. Soc.*, **73**(9), 1365–1374.

Hess, M., P. Koepke and I. Schult, 1998: Optical properties of aerosols and clouds: the software package OPAC. *Bull. Amer. Meteor. Soc.*, **79**(5), 831–844.

van de Hulst, H. C., 1981: *Light Scattering by Small Particles.* Dover Publications. 470 pp.

Jackson, J. D., 1975: *Classical Electrodynamics.* John Wiley & Sons, New York, 2nd edn. 848 pp.

Jaggard, D. L., C. Hill, R. W. Shorthill, D. Stuart, M. Glantz, F. Rosswog, B. Taggart and S. Hammond, 1981: Light scattering from particles of regular and irregular shape. *Atmos. Environ.*, **15**(12), 2511–2519.

Jones, D. M. A., 1959: The shape of raindrops. *J. Meteor.*, **16**, 504–510.

Joss, J. and E. G. Gori, 1978: Shapes of raindrop size distributions. *J. Appl. Meteor.*, **17**, 1054–1061.

Kaula, W. M., 1968: *An introduction to planetary physics.* Space science text series. John Wiley & Sons, New York.

Kubesh, R. J. and K. V. Beard, 1993: Laboratory measurements of spontaneous oscillations for moderate-size raindrops. *J. Atmos. Sci.*, **50**(8), 1089–1098.

Lamberg, L., K. Muinonen, J. Ylönen and K. Lumme, 2001: Spectral estimation of Gaussian random circles and spheres. *J. Comp. Appl. Math.*, **136**, 109–121.

Lehtelä, L., A. K. Piironen and T. Nousiainen, 1999: Scattering model for precipitation type sensor. In: *4th Conference on Electromagnetic and Light Scattering by Nonspherical Particles: Theory and Applications, book of extended abstracts.* Vigo, Spain.

Liou, K.-N., 1980: *An introduction to atmospheric radiation*, vol. 26 of *International geophysics series.* Academic Press. 392 pp.

Lumme, K. and J. Rahola, 1994: Light scattering by porous dust particles in the discrete-dipole approximation. *Astrophys. J.*, **425**, 653–667.

Macke, A. and M. Großklaus, 1998: Light scattering by nonspherical raindrops: Implications for lidar remote sensing of rainrates. *J. Quant. Spectrosc. Radiat. Transfer*, **60**, 355–363.

Marshall, J. S. and W. McK. Palmer, 1948: The distribution of raindrops with size. *J. Meteor.*, **5**, 165–166.

Matson, R. J. and A. W. Huggins, 1980: The direct measurement of the sizes, shapes and kinematics of falling hailstones. *J. Atmos. Sci.*, **37**, 1107–1125.

Mishchenko, M. I. and A. Macke, 1999: How big should hexagonal ice crystals be to produce halos? *Appl. Opt.*, **38**(9), 1626–1629.

Mishchenko, M. I. and L. D. Travis, 1998: Capabilities and limitations of a current Fortran implementation of the T-matrix method for randomly oriented rotationally symmetric scatterers. *J. Quant. Spectrosc. Radiat. Transfer*, **60**, 309–324.

Mishchenko, M. I., A. A. Lacis, B. E. Carlson and L. D. Travis, 1995: Nonsphericity of dust-like tropospheric aerosols: implications for aerosol remote sensing and climate modeling. *Geophys. Res. Lett.*, **22**(9), 1077–1080.

Mishchenko, M. I., L. D. Travis, R. A. Kahn and R. A. West, 1997: Modeling phase functions for dustlike tropospheric aerosols using a shape mixture of randomly oriented polydisperse spheroids. *J. Geophys. Res.*, **102**(D14), 16831–16847.

Mishchenko, M. I., L. D. Travis and A. Macke, 2000a: T-matrix method and its applications. In: *Light Scattering by Nonspherical Particles* (edited by Mishchenko, M. I., J. W. Hovenier and L. D. Travis), chap. 6, pp. 147–172. Academic Press, San Diego.

Mishchenko, M. I., W. J. Wiscombe, J. W. Hovenier and L. D. Travis, 2000b: Overview of scattering by nonspherical particles. In: *Light Scattering by Nonspherical Particles* (edited by Mishchenko, M. I., J. W. Hovenier and L. D. Travis), chap. 2, pp. 29–60. Academic Press, San Diego.

Muñoz, O., H. Volten, J. F. de Haan, W. Vassen and J. W. Hovenier, 2001: Experimental determination of scattering matrices of randomly oriented fly ash and clay particles at 442 and 633 nm. *J. Geophys. Res.*, **106**(D19), 22833–22844.

Mugnai, A. and W. J. Wiscombe, 1980: Scattering of radiation by moderately nonspherical particles. *J. Atmos. Sci.*, **37**, 1291–1307.

Muinonen, K., 1989: Scattering of light by crystals: a modified kirchhoff approximation. *Appl. Opt.*, **28**, 3044–3050.

Muinonen, K., 1990: *Light scattering by inhomogeneous media: backward enhancement and reversal of linear polarization.* Ph.D. thesis, Univ. Helsinki, Helsinki.

Muinonen, K., 1996: Light scattering by Gaussian random particles. *Earth, Moon and Planets*, **72**, 339–342.

Muinonen, K., 1998: Introducing the Gaussian shape hypothesis for asteroids and comets. *Astron. & Astrophys.*, **332**, 1087–1098.

Muinonen, K., 2000a: Light scattering by axially symmetric Gaussian random particles. In: *Light Scattering by Nonspherical Particles: Halifax Contributions* (edited by Videen, G., Q. Fu and P. Chýlek), pp. 91–94.

Muinonen, K., 2000b: Light scattering by stochastically shaped particles. In: *Light Scattering by Nonspherical Particles* (edited by Mishchenko, M. I., J. W. Hovenier and L. D. Travis), chap. 11, pp. 323–352. Academic Press, San Diego.

Muinonen, K., 2002a: Coherent backscattering by absorbing and scattering media. In: *Electromagnetic and Light Scattering by Nonspherical Particles* (edited by Gustafson, B., L. Kolokolova and G. Videen), pp. 223–226. ARL, Adelphi, USA.

Muinonen, K., 2002b: Light-scattering approximations for small irregular particles. In: *Electromagnetic and Light Scattering by Nonspherical Particles* (edited by Gustafson, B., L. Kolokolova and G. Videen), pp. 219–222. ARL, Adelphi, USA.

Muinonen, K. and J. S. V. Lagerros, 1998: Inversion of shape statistics for small solar system bodies. *Astron. & Astrophys.*, **333**, 753–761.

Muinonen, K. and K. Saarinen, 2000: Ray optics approximation for Gaussian random cylinders. *J. Quant. Spectrosc. Radiat. Transfer*, **64**, 201–218.

Muinonen, K., K. Lumme, J. I. Peltoniemi and W. M. Irvine, 1989: Light scattering by randomly oriented crystals. *Appl. Opt.*, **28**, 3051–3060.

Nelson, A. R. and N. R. Gokhale, 1972: Oscillation frequencies of freely suspended water drops. *J. Geophys. Res.*, **77**(15), 2724–2727.

Nousiainen, T., 1997: *Light Scattering by Gaussian, Randomly Oscillating Raindrops.* Phil. Lic. thesis, University of Helsinki. 73 pp.

Nousiainen, T. and K. Muinonen, 2002: Modified ray-optics computations for Saharan particles. In: *Electromagnetic and Light Scattering by Nonspherical Particles* (edited by Gustafson, B., L. Kolokolova and G. Videen), pp. 251–254. ARL, Adelphi, USA.

Nousiainen, T., K. Lumme and K. Muinonen, 1995: Light scattering by quartz particles in the geometric optics approximation. In: *Proceedings of the Finnish Astronomical Society* (edited by Muinonen, K. and V. Forsström), pp. 18–21. Helsinki, Finland.

Oguchi, T., 1983: Electromagnetic wave propagation and scattering in rain and other hydrometeors. *Proc. IEEE*, **71**(9), 1029–1078.

Patterson, E. M., D. A. Gillette and B. H. Stockton, 1977: Complex index of refraction between 300 and 700 nm for Saharan aerosols. *J. Geophys. Res.*, **82**(21), 3153–3160.

Peltoniemi, J., T. Nousiainen and K. Muinonen, 1998: Light scattering by Gaussian random particles using various volume-integral-equation techniques. In: *Conference on Light Scattering by Nonspherical Particles: Theory, Measurements, and Applications, Book of Extended Abstracts*, pp. 195–198. AMS, New York, U.S.A.

Peltoniemi, J. I., 1996: Variational volume integral equation method for electromagnetic scattering by irregular grains. *J. Quant. Spectrosc. Radiat. Transfer*, **55**(5), 637–647.

Peltoniemi, J. I., K. Lumme, K. Muinonen and W. M. Irvine, 1989: Scattering of light by stochastically rough particles. *Appl. Opt.*, **28**, 4088–4095.

Pilinis, C. and X. Li, 1998: Particle shape and internal inhomogeneity effects on the optical properties of tropospheric aerosols of relevance to climate forcing. *J. Geophys. Res.*, **103**(D4), 3789–3800.

Pruppacher, H. R. and J. D. Klett, 1997: *Microphysics of Clouds and Precipitation.* Kluwer Academic Publishers, 2nd edn. 954 pp.

Ray, P. S., 1972: Broadband complex refractive indices of ice and water. *Appl. Opt.*, **11**(8), 1836–1844.

Rayleigh, L., 1879: On the capillary phenomena of jets. *Proc. Roy. Soc. London*, **29**, 71–97.

Sauvageot, H., 1994: Rainfall measurement by radar: a review. *Atmospheric Research*, **35**, 27–54.

Schiffer, R., 1989: Light scattering by perfectly conducting statistically irregular particles. *J. Opt. Soc. Amer. A*, **6**(3), 385–402.

Schiffer, R., 1990: Perturbation approach for light scattering by an ensemble of irregular particles of arbitrary material. *Appl. Opt.*, **29**(10), 1536–1550.

Sihvola, A. H., 1989: Self-consistency aspects of dielectric mixing theories. *IEEE Trans. Geosci. Remote Sens.*, **27**(4), 403–415.

Tokay, A. and K. V. Beard, 1996: A field study of raindrop oscillations. Part I: Observation of size spectra and evaluation of oscillation causes. *J. Appl. Meteor.*, **35**, 1671–1687.

Trinh, E., A. Zwern and T. G. Wang, 1982: An experimental study of small-amplitude drop oscillations in immiscible liquid systems. *J. Fluid Mech.*, **115**, 453–474.

Videen, G. W., 1992: *"Light scattering from a sphere on or near an interface.* Ph.D. thesis, University of Arizona. 130 pp.

Volten, H., O. Muñoz, J. F. de Haan, W. Vassen, J. W. Hovenier, K. Muinonen and T. Nousiainen, 2001: Scattering matrices of mineral aerosol particles at 441.6 nm and 632.8 nm. *J. Geophys. Res.*, **106**(D15), 17375–17401.

Warren, S. G., 1984: Optical constants of ice from the ultraviolet to the microwave. *Appl. Opt.*, **23**(8), 1206–1225.

Waterman, P. C., 1971: Symmetry, unitarity, and geometry in electromagnetic scattering. *Phys. Rev. D*, **3**(4), 825–839.

Yang, P. and K. N. Liou, 1998: Single-scattering properties of complex ice crystals in terrestrial atmosphere. *Contrib. Atmos. Phys.*, **71**(2), 223–248.

Yang, P. and K. N. Liou, 2000: Finite difference time domain method for light scattering by nonspherical and inhomogeneous particles. In: *Light Scattering by Nonspherical Particles* (edited by Mishchenko, M. I., J. W. Hovenier and L. D. Travis), chap. 7, pp. 173–221. Academic Press, San Diego.

A Errata

- In Paper I, Eq. (11) should read:

$$C_s(\gamma) = \exp\left(-\frac{2}{l_c^2}\sin^2\frac{1}{2}\gamma\right)$$
$$l_c = 2\sin(\frac{1}{2}\Gamma)$$

- In Paper II, page 648, Fig. 1, vectors $\boldsymbol{e}_l$ and $\boldsymbol{e}_r$ are erroneously indicated as components parallel and perpendicular to the scattering plane, when in fact they refer to the reference spherical coordinate system and should be written $\boldsymbol{e}_\vartheta$ and $\boldsymbol{e}_\varphi$, respectively. The results are not affected.

- In Paper III, page 794, 17th line below Eq. (20), the maximum amplitude of $A_{2,0}$ should be 0.05.

- In Paper IV, page 649, 9th line from the bottom: 0.0059 should be 0.059.