# LIGHT SCATTERING BY SMALL PARTICLES

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# PREFACE

The scattering of electromagnetic waves by a homogeneous sphere is a problem with a known solution. I first met this problem when I needed some numbers and curves in an astrophysical investigation. I soon learned that it is a long way from the formulae containing the solution to reliable numbers and curves. Subsequent conversations and correspondence with other research workers, notably in chemistry, showed that the same difficulty was felt in other fields.

The studies on which the present book is based were started in 1945 in an attempt to compile the data available in the literature and to fill in the gaps, where needed. Several related problems, such as the scattering by cylinders, were added to the original topic.

For clearer presentation, the problems of mathematical physics dealing with the scattering properties of single particles (part II) have been separated from the problems arising in specific fields of application (part III). The properties of the particles that should be known in order to describe the optical properties of a medium consisting of such particles have been defined in general terms (part I).

New formulae or numerical results are contained in almost all chapters. They are noted in the references at the end of each chapter. The reference lists have steadily grown in the course of the years; they probably are fairly complete, but no systematic bibliographical study has been made.

Although the book has a mathematical character, requirements of mathematical rigor do not dominate the presentation. Arguments based on physical intuition are given wherever they illuminate the subject more clearly than a mathematical derivation. Simple results that arise under special sets of assumptions are often derived both ways. In view of the wishes expressed by several colleagues, I have not shrunk from a certain inconsistency in the level of presentation. For instance, chapter 17, which comes closest to an actual research report, contains less explanation of elementary detail than some earlier chapters, which may be consulted by research workers without special mathematical training.

Personal acknowledgment of the support received from numerous friends and colleagues in writing this book is impossible. I wish to express my thanks both for the information they have contributed and for their inspiring questions.

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*Leiden, The Netherlands*
*March, 1957*

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# PART I

# Basic Scattering Theory

# 1. INTRODUCTION

## 1.1. Scattering, Absorption, Extinction

This book is a treatise on the *scattering* of light. Hardly ever is light observed directly from its source. Most of the light we see reaches our eyes in an indirect way. Looking at a tree, or a house, we see diffusely reflected sunlight. Looking at a cloud, or at the sky, we see scattered sunlight. Even an electric lamp does not send us light directly from the luminous filament but usually shows only the light that has been scattered by a bulb of ground glass. Everyone engaged in the study of light or its industrial applications meets the problem of scattering.

Scattering is often accompanied by *absorption*. A leaf of a tree looks green because it scatters green light more effectively than red light. The red light incident on the leaf is absorbed; this means that its energy is converted into some other form (what form of energy is irrelevant for our purpose) and is no longer present as red light. Absorption is preponderant in materials such as coal and black smoke; it is nearly absent (at visual wavelengths) in clouds.

Both scattering and absorption remove energy from a beam of light traversing the medium: the beam is attenuated. This attenuation, which is called *extinction*, is seen when we look directly at the light source. The sun, for instance, is fainter and redder at sunset than at noon. This indicates an extinction in the long air path, which is strong in all colors but even stronger in blue light than in red light. Whether scattering or absorption is mainly responsible for this extinction cannot be judged from this observation alone. Looking sideways at the air, through which the sun shines, we see that actually blue light is scattered more strongly. Measurements show that all light taken away from the original beam reappears as scattered light. Therefore, scattering, and not absorption, causes the extinction in this example.

Other terminology is sometimes used but is not recommended. Here the word absorption is used in the sense of extinction as defined above.[1] Actual absorption is then designated as "pure absorption" or "true absorption." Throughout this book terms will be used as defined above, so that

$$\text{Extinction} = \text{scattering} + \text{absorption}.$$

[1] E.g., in the term "interstellar absorption."

## 1.2. Subject Limitations

Only a few of the multitude of scattering phenomena are treated in this book.

A first restriction is that we shall always assume that the scattered light has the same frequency (i.e., the same wavelength) as the incident light[2]. Effects like the Raman effect, or generally any quantum transitions, are excluded.

### 1.21 Independent Scattering

A second, most important limitation is that independent particles are considered. The distinction is roughly this: the scattering by well-defined separate particles, such as occur in a fog, is within the province of this book, whereas the scattering by a diffuse medium, as for instance a solution of a high polymer, is not discussed.

A more precise distinction may be made. If light traverses a perfectly homogeneous medium, it is not scattered. Only inhomogeneities cause scattering. Now, in fact, any material medium has inhomogeneities as it consists of molecules, each of which acts as a scattering center, but it depends on the arrangement of these molecules whether the scattering will be very effective. In a perfect crystal at zero absolute temperature the molecules are arranged in a very regular way, and the waves scattered by each molecule interfere in such a way as to cause no scattering at all but just a change in the overall velocity of propagation. In a gas, or fluid, on the other hand, statistical fluctuations in the arrangement of the molecules cause a real scattering, which sometimes may be appreciable. In these examples, whether or not the molecules are arranged in a regular way, the final result is a *cooperative effect* of all molecules. The scattering theory then has to investigate in detail the phase relations between the waves scattered by neighboring molecules. Any such problem, in which the major difficulty is in the precise description of the cooperation between the particles, is called a problem of dependent scattering and is not treated in this book.[3]

Frequently, however, the inhomogeneities are alien bodies immersed in the medium. Obvious examples are water drops and dust grains in atmospheric air and bubbles in water or in opal glass. If such particles are sufficiently far from each other, it is possible to study the scattering

[2] This may technically be called coherent scattering. However, this term is often used with a different connotation: an assembly of particles is said to scatter incoherently if the positions of the individual particles vary sufficiently (sec. 1.21).

[3] See the references at the end of this chapter. References appear throughout at the end of each chapter.

by one particle without reference to the other ones. This will be called *independent scattering*; it is the exclusive subject of this book.

It may be noted that waves scattered by different particles from the same incident beam in the same direction still have a certain phase relation and may still interfere. The fact that the wavelength remains the same means that the scattered waves must be either in phase and enhance each other or out of phase and destroy each other, or any intermediate possibility. The assumption of independent scattering implies that there is no systematic relation between these phases. A slight displacement of one particle or a small change in the scattering angle may change the phase differences entirely. The net effect is that *for all practical purposes the intensities scattered by the various particles must be added* without regard to phase. It thus seems that the scattering by different particles is incoherent, although in the strict sense this is not true. An exception must be made for virtually zero scattering angles. In these directions no scattering in the ordinary sense can be observed. (See chap. 4.)

What distance between particles is sufficiently large to ensure independent scattering? Early estimates have shown that a mutual distance of 3 times the radius is a sufficient condition for independence. This may not be a general rule, but a more precise discussion is beyond the scope of this book. In most practical problems the particles are separated by much larger distances. Even a very dense fog consisting of droplets 1 mm in diameter and through which light can penetrate only 10 meters has about 1 droplet in 1 $cm^3$, which means that the mutual distances are some 20 times the radii of the drops. The same is true for many colloidal solutions.

### 1.22. Single Scattering

A third limitation is that the effects of multiple scattering will be neglected. Practical experiments most often employ a multitude of similar particles in a cloud or a solution. The obvious relations for a thin and tenuous cloud containing $M$ scattering particles are that the intensity scattered by the cloud is $M$ times that scattered by a single particle, and the energy removed from the original beam (extinction) is also $M$ times that removed by a single particle. This simple proportionality to the number of particles holds only if the radiation to which each particle is exposed is essentially the light of the original beam.

Each particle is also exposed to light scattered by the other particles, whereas the light of the original beam may have suffered extinction by the other particles. If these effects are strong, we speak of *multiple scattering* and a simple proportionality does not exist. This situation may be illustrated by a white cloud in the sky. Such a cloud is like a dense fog; its droplets may be considered as independent scatterers. Yet the total intensity scattered by the cloud is not proportional to the number of droplets contained in it, for not each droplet is illuminated by full sunlight. Drops within the cloud may receive no direct sunlight at all but only diffuse light which has been scattered by other drops. Most of the light that emerges from a cloud has been scatttered by two or more droplets successively. It is estimated (for a very thick cloud) that about 10 per cent emerges after a single scattering.

Multiple scattering does not involve new physical problems, for the assumption of independence, which states that each droplet may be *thought to be in free space, exposed to light from a distant source, holds* true whether this source is the sun or another droplet. Yet the problem of finding the intensities inside and outside the cloud is an extremely difficult mathematical problem. This problem has been studied extensively in many ramifications. It is usually called the problem of radiative transfer. Common applications are the transfer of radiation in a stellar atmosphere and the scattering of neutrons in an atomic pile. The cases treated so far refer to rather simple forms both of the single scattering pattern (isotropic scattering, Rayleigh scattering) and of the entire cloud (infinite or finite slab with plane boundaries, sphere). The reader is referred to the literature for further details.

A simple and conclusive test for the absence of multiple scattering is to double the concentrations of particles in the investigated sample. If the scattered intensity is doubled, only single scattering is important. Another criterion may be the extinction. The intensity of a beam passing through the sample is reduced by extinction to $e^{-\tau}$ of its original value. Here $\tau$ is the optical depth of the sample along this line. If $\tau < 0.1$ single scattering prevails; for $0.1 < \tau < 0.3$ a correction for double scattering may be necessary. For still larger values of the optical depth the full complexities of multiple scattering become a factor. They may not prevent a determination of the scattering properties of a single particle, but they certainly make the interpretation much less clear. Caution is invariably required when the optical depth is not small in all directions through the sample.

Concluding this section it may be noted that this book treats only the very simplest case occurring in the theory of many particles. This leaves room for a thorough treatment of the scattering theory for one particle.

## 1.3. Historical Review

A proper understanding of the subject will be helped greatly by a review of its history, even though this has to be brief and can only show some of the highlights.