Absorption and Scattering of Light by Small Particles

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Library of Congress Card No.:
Applied for

British Library Cataloging-in-Publication Data:
A catalogue record for this book is available from the British Library

Bibliographic information published by
Die Deutsche Bibliothek
Die Deutsche Bibliothek lists this publication in the Deutsche Nationalbibliografie; detailed bibliographic data is available in the Internet at <http://dnb.ddb.de>.

Wiley Professional Paperback Edition Published 1998

Printed in the Federal Republic of Germany
Printed on acid-free paper

Printing Strauss GmbH, Mörlenbach
Bookbinding Litges & Dopf Buchbinderei GmbH, Heppenheim

ISBN-13: 978-0-471-29340-8
ISBN-10: 0-471-29340-7

To our families

Preface

When asked during the writing of this book what topic could divert us for so long from the pleasures of a normal life we would answer: "It is about how small particles absorb and scatter light." "My goodness," would be the response, "who could possibly be interested in that?" As it happens, scientists and engineers from a surprising variety of disciplines—solid-state physics, electrical engineering, meteorology, chemistry, biophysics, astronomy—make forays into this field, some never to escape. To completely satisfy such diverse groups, each with its peculiar conventions, notation, terminology, and canons, is an impossible task: physicists prefer the language of elementary excitations—phonons, plasmons, and all that; electrical engineers are more comfortable with the language of antennas and waveguides; chemists and biophysicists might not like either. We have therefore striven for the middle ground with the hope of, if not pleasing everyone, at least not antagonizing anyone. Ultimately, however, our point of view is that of physicists. Quantum-mechanical concepts are introduced where they serve to elucidate physical phenomena, but otherwise our approach is primarily classical.

Like so many other books, this one began its existence as lecture notes. Separately and jointly we have given lectures to graduate students and researchers with the kinds of diverse backgrounds and interests we expect our readers will have. Although more of an advanced monograph than a textbook, this book has a pedagogical flavor because of its origins. Indeed, many of the topics covered are in direct response to questions asked either in classrooms or by our colleagues.

There is one important idea, the *raison d'être* of this book, that we should like to implant firmly in the minds of our readers: scattering theory divorced from the optical properties of bulk matter is incomplete. Solving boundary-value problems in electromagnetic theory may be great fun and often requires considerable skill; but the full physical ramifications of mathematical solutions are hidden to those with little knowledge of how refractive indices of various solids and liquids depend on frequency, the values they take, and the constraints imposed on them. Accordingly, this book is divided into three parts.

Part 1, Chapters 1 through 8, is primarily scattering theory. After an introduction there is a chapter on those topics from electromagnetic theory essential to an understanding of the succeeding six chapters on exact and approximate solutions to various scattering problems. Because uninterrupted strings of mathematical formulas tend to pall, computational and experimental results are interspersed throughout these chapters.

Bulk matter, rather than particles, is the subject of Part 2. In Chapter 9 we discuss classical theories of optical properties based on idealized models. Such models rarely conform strictly to reality, however, so Chapter 10 presents measurements for three representative materials over a wide range of frequencies, from radio to ultraviolet: aluminum, a metal; magnesium oxide, an insulator; and water, a liquid.

Part 3 is a marriage of Parts 1 and 2, the offspring of which are chapters on extinction (Chapter 11), surface modes (Chapter 12), and angular scattering (Chapter 13). Applications are not totally absent from the first thirteen chapters, but there is a greater concentration of them in Chapter 14.

We did not attempt an exhaustive list of references, even assuming that were possible. Instead, we concentrated on the years since publication of Kerker's book (1969), which cites nearly a thousand references. Even with this restriction we were selective, guided by our tastes rather than some ideal notion of completeness.

We avoided irritating statements such as "it can be shown"; while implying calm, they usually signal rough sailing ahead. Of course, we do not give all the details of lengthy derivations, but we do provide enough guideposts so that a reader can, with a bit of effort, duplicate our results. We always chose the simplest derivations, preferring physical plausibility over mathematical rigor. Those who demand the latter are reminded that one man's rigor is another man's mortis.

This book was not written with scissors: all derivations are our own, as are most of the figures, many of them generated with the computer programs in the appendixes. Even much of the experimental data was taken with an eye toward examples for the book. Any errors, therefore, are solely ours.

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January 1983

Acknowledgments

During much of the writing of this book I was a wandering scholar. At each institution I visited I widened the circle of those to whom I am indebted for suggestions, comments, and encouragement. Although fading memory prevents me from adequately expressing my gratitude to all of them, there are many whose contributions remain fixed in my mind.

Daya Gilra, my office-mate in the Department of Applied Mathematics and Astronomy at University College, Cardiff, Wales, suggested that I give a course of lectures on light scattering, the notes for which subsequently formed some of the raw material for this book. For this suggestion and for much more, I am grateful. My thanks also go to those who faithfully attended these lectures, particularly Harry Abadi and Indra Dayawansa, my collaborators, and Joachim Köppen. I would be remiss if I did not acknowledge the assistance of two members of the Pure Mathematics-Department at Cardiff, W. D. (Des) Evans and George Greaves.

Louis Battan provided me with a haven for over a year in the Institute of Atmospheric Physics at the University of Arizona, and his support of my endeavors has never flagged, although I have not always followed his sage advice. Sean Twomey was an incisive critic, a fertile source of ideas, and an arbiter of disputes. Without his constant goading—"how many pages did you write today, Craig?"—the writing of this book would have continued into the hereafter. Margaret Sanderson Rae patiently answered hundreds of questions about matters of style and scrutinized some of the first chapters to keep me from straying too far from good usage. E. Philip Krider and Michael Box also read parts of the manuscript, and I thank them for their suggestions.

At Arizona I was partly supported by the Department of Physics through the generosity of Robert Parmenter. I am also indebted to other members of this department, particularly John Kessler, Michael Scadron, Bernard Bell, Rein Kilkson, and William Bickel.

A grant from the Institute of Occupational and Environmental Health in Montreal, obtained through the kind assistance of George Wright, enabled me to return to Wales, where I worked with Vernon Timbrell in the Medical Research Council Pneumoconiosis Unit at Llandough Hospital. It was there that most of Chapter 8 was written as well as the first version of Appendix C.

Further work on this book was undertaken at Los Alamos Scientific Laboratory, for which I must thank Paul Mullaney. I am also grateful to Gary Salzman, who helped in many ways, and to Sally Wilkins, who inspected the programs in the appendixes and made several suggestions for improving them; she is not, however, responsible for the numbers they produce.

My colleagues at Pennsylvania State University, Alistair Fraser, John Olivero, and Timothy Nevitt, are to be thanked for helping me to keep this book as free from errors as possible.

Others who deserve a word of thanks are Alfred Holland and Arlon Hunt.

At Wiley, Beatrice Shube, in the words of the Beatles, "was like a mum to us."

My gratitude is deepest, however, to Nanette Malott Bohren, who followed me without complaint on my wanderings, was neglected for over three years, but who nevertheless read every page of the manuscript—several times—thereby improving its readability.

C.F.B.

For helping me to learn about the interaction of light with small particles I sincerely thank the other students and co-workers who have been my colleagues over the years: James L. Stapp, Terry Steyer, Roger Perry, Janice Rathmann, Otto Edoh, Lin Oliver, Wolfgang Krätschmer, and Kenrick Day. Special thanks are due to Arlon Hunt for the work we shared in the days when everything about small particles was new to us, exciting, and occasionally explosive.

D.R.H.

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Part 1

Basic Theory

Chapter 1

Introduction

Cumulus clouds in the summer afternoon sky present a striking contrast of white against a bright blue sky. During a sudden thundershower the primary and secondary rainbows display their multicolored arches. Other colors in nature are the dark green of forest foliage and the red and orange hues of the Grand Canyon in early morning. High in the mountains or on the desert when the air is clean one can clearly see dark patches in the bright band of the Milky Way. Chimney smut turns all it touches to dirty blackness, and iridescent opal shimmers with a variety of colors. All these visual phenomena and many more are manifestations of scattering and absorption of light by small particles, which is the subject of this book. We do not, however, restrict ourselves to visible light.

The study of light scattering and its applications is an enormous field, much too large to be treated successfully in one book. We limit our treatment, therefore, to scattering by single particles within the framework of classical electromagnetic theory and linear optics. Not all the examples given above fall within these restrictions. The colors of rocks and foliage, for example, involve complicated interactions of light with many densely packed centers of scattering and absorption. Nevertheless, all detailed treatments of the more complicated phenomena begin with those that we shall study.

1.1 PHYSICAL BASIS FOR SCATTERING AND ABSORPTION

Scattering of electromagnetic waves by any system is related to the heterogeneity of that system: heterogeneity on the molecular scale or on the scale of aggregations of many molecules. Regardless of the type of heterogeneity, the underlying physics of scattering is the same for all systems. Matter is composed of discrete electric charges: electrons and protons. If an obstacle, which could be a single electron, an atom or molecule, a solid or liquid particle, is illuminated by an electromagnetic wave (Fig. 1.1), electric charges in the obstacle are set into oscillatory motion by the electric field of the incident wave. Accelerated electric charges radiate electromagnetic energy in all directions; it is this *secondary radiation* that is called the radiation *scattered* by the

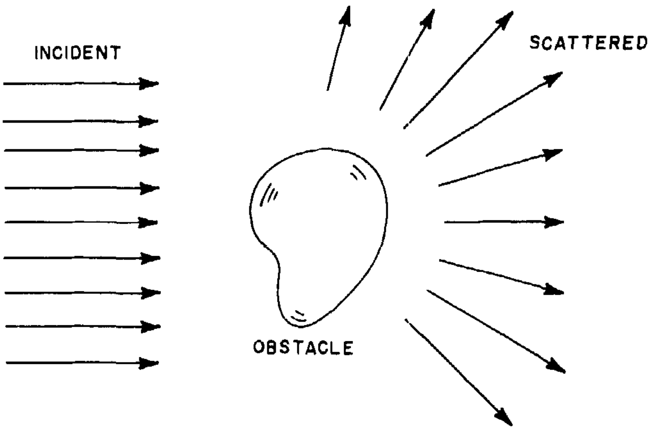

Figure 1.1 Scattering by an obstacle.

obstacle:

$$\text{Scattering} = \text{excitation} + \text{reradiation}$$

In addition to reradiating electromagnetic energy, the excited elementary charges may transform part of the incident electromagnetic energy into other forms (thermal energy, for example), a process called *absorption*. Scattering and absorption are not mutually independent processes, and although, for brevity, we often refer only to scattering, we shall always mean absorption as well.

1.2 SCATTERING BY FLUCTUATIONS AND BY PARTICLES

Everything except a vacuum is heterogeneous in some sense. Even in media that we usually consider to be homogeneous (e.g., pure gases, liquids, or solids) it is possible to distinguish the individual heterogeneities (atoms and molecules) with a sufficiently fine probe. Therefore, all media scatter light. In fact, many phenomena that are not usually referred to as scattering phenomena are ultimately the result of scattering. Among these are: (1) diffuse reflection by rough surfaces; (2) diffraction by slits, gratings, and edges; and (3) specular reflection and refraction at optically smooth interfaces. Let us examine the third example in more detail. The directions of the reflected and refracted rays in Fig. 1.2 are specified by the law of specular reflection and Snell's law. These laws have been known empirically for a long time. They can, however, be derived by using the Maxwell equations, and in so doing one also obtains the amplitudes and phases of the reflected and refracted rays (i.e., the Fresnel equations). It is also possible, although far from easy, to derive these laws by explicitly considering the molecular nature of matter. The transparent medium

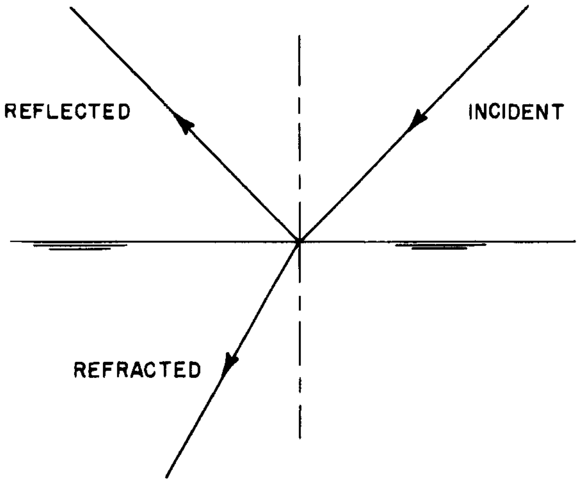

Figure 1.2 Reflection and refraction at an optically smooth interface.

on which light is incident (Fig. 1.2) is an aggregation of very many molecules. The field in the neighborhood of a given molecule induces an oscillating dipole moment in the molecule, which in turn gives rise to secondary dipole radiation. Solids, liquids, and many gases are *optically dense*: the molecular separation is much less than the wavelength of the incident light. In solids and liquids the molecular separation is about 2–3 Å, whereas for gases at standard temperature and pressure (STP) the average separation is about 30 Å. Thus, each molecule is acted on not only by the incident field but also by the resultant of the secondary fields of all the other molecules. But the secondary field of a molecule depends on the field to which it is exposed. Therefore, we have an electromagnetic many-body problem: the molecules are coupled. The net result of the solution to this problem, subject to suitable approximations, is that inside the medium the secondary waves superpose on each other and on the incident wave to give just a refracted wave with propagation velocity c/n, where c is the speed of light *in vacuo* and n is the refractive index. The incident wave is completely extinguished inside the medium; this is called the Ewald–Oseen extinction theorem. Outside the medium the secondary waves superpose to give a specularly reflected wave. As we shall see in Chapter 9, n depends on the number of molecules per unit volume and the polarizability of a single molecule; this underscores the assertion that refraction is a scattering phenomenon in its essentials: the refractive index is a manifestation of scattering by the many molecules that comprise the medium.

Careful observation of the configuration shown in Fig. 1.2 reveals a phenomenon that contradicts the familiar law of refraction. Suppose that we completely darken the surroundings and illuminate a transparent medium, which could be pure water, with an intense laser beam. Even if the medium is free from all particulate contamination, the path of the beam in the medium can be dimly perceived (this might require a suitable detector) if we look in directions other than in the plane of incidence. This is not compatible with Snell's law, which asserts that the refracted ray lies in the plane of incidence and which takes no account of light other than in this plane. Therefore, Snell's law is only a first approximation, and we must probe deeper to discover the origins of this light weakly scattered in all directions, which is superposed on the more intense unidirectional refracted beam.

It is assumed in the usual analysis of the interaction of a beam of light with an optically smooth interface that the refracting medium is perfectly homogeneous, whereas, in fact, it is only *statistically homogeneous*. That is, the average number of molecules in a given volume element is constant, but at any instant the number of molecules in this element will be different compared with any other instant. It is these *density fluctuations* which give rise to scattering in optically dense media. Although we, and others, refer to scattering *by* fluctuations for brevity, we must emphasize that molecules are the scattering agents. But one can ignore them and imagine that their scattering is the result of local density fluctuations in an otherwise homogeneous medium. It would be more precise, therefore, to refer to the fluctuation theory of scattering by molecules rather than to scattering by fluctuations.

There are other types of fluctuations. For example, if sugar is dissolved in water, after thorough stirring the sugar concentration will be statistically homogeneous, but *concentration fluctuations* will give rise to scattering. If the molecules are nonspherical, there will be *orientation fluctuations*.

All such scattering by fluctuations is excluded here; we confine ourselves to scattering by particles, and a fluctuation is not a particle in our sense. It is important to distinguish between scattering by fluctuations and scattering by particles because there is a large body of literature on "light scattering" which is clearly distinct from the subject of this book. Although the mathematical expressions are often similar, the underlying physics is somewhat different: scattering by fluctuations, for example, involves thermodynamic arguments, whereas scattering by particles does not. Moreover, there is common terminology, which is a possible source of confusion. For example, scattering by density fluctuations in ideal gases has the same functional form as scattering by dilute suspensions of particles small compared with the wavelength. We shall call the latter type of scattering Rayleigh scattering, but in the theory of scattering by fluctuations the term may have a somewhat different meaning. We refer the reader to an article by Young (1982) in which he attempts to sort out all the different ways in which "Rayleigh scattering" is used and misused.

The basic problem to which we confine our attention is the interaction of light of arbitrary wavelength with a single particle (i.e., a well-defined aggregate of very many atoms or molecules), which is embedded in an otherwise homogeneous medium (Fig. 1.3). By homogeneous is meant that the atomic or molecular heterogeneity is small compared with the wavelength of the incident light; we also ignore scattering by fluctuations, which is usually much less than

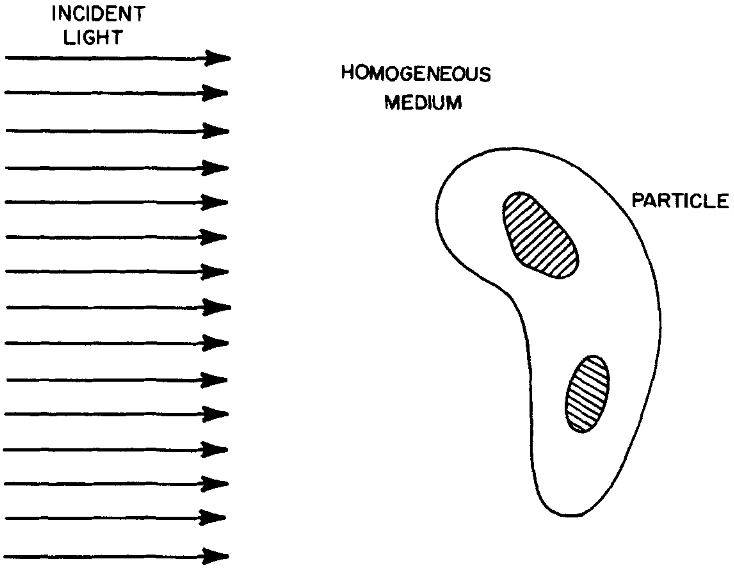

Figure 1.3 Interaction of light with a single particle.

scattering by particles. Although the particle may be complicated in shape and may have several homogeneous components, we assume that it is composed of matter that is at every point describable in macroscopic terms. That is, the optical properties of a particle or regions thereof are completely specified by frequency-dependent optical constants; the interaction of photons with elementary quantum excitations need not be considered explicitly.

We restrict our treatment to *elastic* scattering: the frequency of the scattered light is the same as that of the incident light. This excludes from study *inelastic* scattering phenomena such as Mandel'stam–Brillouin and Raman scattering. Elastic scattering is sometimes denoted as *coherent* scattering, but elastic is more physically descriptive and the notion of coherence as a definite phase relation between different sources of radiation is firmly established in optics. To add to the confusion, Rayleigh scattering is sometimes used to denote scattering in which there is no change in frequency. Again, see the article by Young (1982) for clarification.

1.3 PHYSICS OF SCATTERING BY A SINGLE PARTICLE

We can acquire a qualitative understanding of the physics of scattering by a single particle without invoking a specific particle or doing any computations. Consider an arbitrary particle, which we conceptually subdivide into small regions (Fig. 1.4). An applied oscillating field (e.g., an incident electromagnetic wave) induces a dipole moment in each region. These dipoles oscillate at the frequency of the applied field and therefore scatter secondary radiation in all

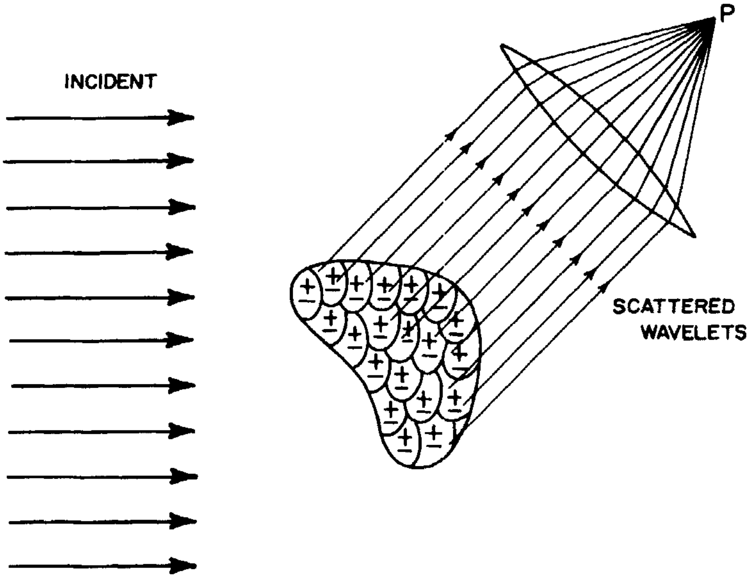

Figure 1.4 The total scattered field at P is the resultant of all the wavelets scattered by the regions into which the particle is subdivided.

directions. In a particular direction (i.e., at a distant point P), the total scattered field is obtained by superposing the scattered wavelets, where due account is taken of their phase differences: scattering by the dipoles is coherent. In general, these phase relations change for a different scattering direction; we therefore expect the scattered field to vary with scattering direction. If the particle is small compared with the wavelength, all the secondary wavelets are approximately in phase; for such a particle we do not expect much variation of scattering with direction. That this is indeed so will be shown in Chapter 5. As the particle size is increased, however, the number of possibilities for mutual enhancement and cancellation of the scattered wavelets increases. Thus, the larger the particle, the more peaks and valleys in the scattering pattern. Shape is also important: if the particle in Fig. 1.4 is distorted, all the phase relations, hence the scattering pattern, are different.

The phase relations among the scattered wavelets depend on geometrical factors: scattering direction, size, and shape. But the amplitude and phase of the induced dipole moment for a given frequency depend on the material of which the particle is composed. Thus, for a full understanding of scattering and absorption by small particles, we need to know how bulk matter responds to oscillatory electromagnetic fields; this is the subject of Chapters 9 and 10.

Methods for calculating scattering by particles are physically equivalent to the procedure outlined above, although their mathematical form may obscure the underlying physics. For certain classes of particles, however, the scattered field may be approximated by subdividing the particle into dipole scatterers and superposing the scattered wavelets; this is what is done in the Rayleigh–Gans approximation (Chapter 6), where interactions among the dipoles are ignored. A more general computational technique in which interactions among the dipoles are accounted for is that of Purcell and Pennypacker (1973) (see Section 8.6).

1.4 COLLECTIONS OF PARTICLES

The fundamental problem under consideration is scattering and absorption by single particles; in natural environments, however, we are usually confronted with collections of very many particles. Even in the laboratory, where it is possible to do experiments with single particles, it is more usual to make measurements on many particles. A rigorous theoretical treatment of scattering by many particles is indeed formidable (see, e.g., Borghese et al., 1979). But if certain conditions are satisfied, a collection poses no more analytical problems than does a single isolated particle.

Particles in a collection are electromagnetically coupled: each particle is excited by the external field and the resultant field scattered by all the other particles; but the field scattered by a particle depends on the total field to which it is exposed. Considerable simplification results if we assume *single scattering*: the number of particles is sufficiently small and their separation sufficiently large that, in the neighborhood of any particle, the total field scattered by all the particles is small compared with the external field. With this assumption the total scattered field is just the sum of the fields scattered by the individual particles, each of which is acted on by the external field in isolation from the other particles. It is difficult to state precise general conditions under which the single scattering criterion is satisfied; it is not satisfied, for example, by clouds, where *multiple scattering* can be appreciable. In laboratory experiments, however, it is usually possible to prepare dilute suspensions of sufficiently small size to ensure single scattering.

We shall assume, in addition to single scattering, that the particles are many and their separations random, which implies *incoherent scattering*. That is, there is no systematic relation among the phases of the waves scattered by the individual particles; thus, the total irradiance scattered by the collection is just the sum of the irradiances scattered by the individual particles. Even, however, in a collection of randomly separated particles, the scattering is coherent in the forward direction, a subject to which we shall return in Chapter 3.

1.5 THE DIRECT AND INVERSE PROBLEM

There are two general classes of problems in the theory of the interaction of an electromagnetic wave with a small particle.

The Direct Problem. Given a particle of specified shape, size, and composition, which is illuminated by a beam of specified irradiance, polarization, and

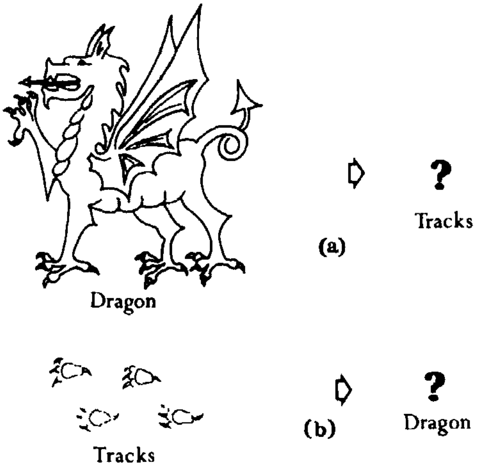

Figure 1.5 (*a*) The direct problem: Describe the tracks of a given dragon. (*b*) The inverse problem: Describe a dragon from its tracks.

frequency, determine the field everywhere. This is the "easy" problem; it consists of describing the tracks of a given dragon (Fig. 1.5*a*).

The Inverse Problem. By a suitable analysis of the scattered field, describe the particle or particles that are responsible for the scattering. This is the "hard" problem; it consists of describing a dragon from an examination of its tracks (Fig. 1.5*b*).

Unfortunately, the problem that is most frequently of interest is the inverse problem, the type of problem to which Sherlock Holmes might have directed his powers had he been a physical scientist rather than a detective. For example, the composition of interstellar dust is one of the major unsolved mysteries in astronomy. Although there is little doubt about the existence of this dust, it has defied complete identification despite considerable effort. The only means for its investigation is analysis of light of various wavelengths that traverses the dust without undergoing scattering or absorption and, less commonly, the light scattered in various directions by the dust. In laboratory investigations, light scattering techniques are often used to determine the size of particles of known shape and composition. Radar backscattering can be used to discriminate between rain and hail. In all these applications the basic problem is to describe a dragon (or flock of dragons) by detailed examination of its tracks. To understand why we have labeled this the "hard" problem, consider that the information necessary to specify a particle uniquely is (1) the vector amplitude and phase of the field scattered in all directions, and (2) the field inside the particle (Hart and Gray, 1964). The field inside a particle is not usually accessible to direct measurement, although under certain conditions, which are not likely to be met except in the laboratory, this field can be approximated by the incident field (see Chapter 6). Even in this special case, however, the amplitude and phase of the scattered field are required; although this is not impossible in principle, it is rarely achieved in practice. The measurements usually available for analysis are the irradiance of the scattered light for a set of directions. We are therefore almost always faced with the task of trying to describe a particle (or worse yet, a collection of particles) with a less than theoretically ideal set of data in hand. But this is not necessarily cause for despair. Often, supplementary information about the particles, some of which is obtained by means other than light scattering techniques, is sufficient to enable them to be described. Thus, we should not be too ready to discard what little information is available or might become available. One source of information sometimes neglected is the polarization properties of the scattered light, a subject we treat in detail in succeeding chapters.

NOTES AND COMMENTS

An excellent concise treatment of scattering—by molecules and particles, single and multiple—at an intermediate level is Chapter 14 of Stone (1963). Among the books devoted entirely to scattering by particles, that by Shifrin (1951) most closely resembles ours in that it discusses optical properties of bulk matter as well. But the two books that have influenced us most are those of van de Hulst (1957) and Kerker (1969); we are indebted to both authors. Another book on scattering, which emphasizes polydispersions, is by Deirmendjian (1969).

There are also more limited treatments of scattering. McCartney (1976, Chaps. 4–6) confines his attention to scattering by atmospheric particles. This is also discussed by Twomey (1977, Chaps. 9–10) in his treatise on atmospheric aerosols. In Goody (1964, Chap. 7) there are discussions of absorption by gases and, in less detail, extinction by molecules and by droplets. Parts of books on electromagnetic theory or optics include the theory of scattering by a sphere, most notably Stratton (1941, pp. 563–573) and Born and Wolf (1965, pp. 633–664). The latter also derive the Ewald–Oseen extinction theorem and apply it to reflection and refraction at a plane interface (pp. 98–104).

The fluctuation theory of scattering by molecules is treated in books by Bhagavantam (1942), Fabelinskii (1968), and Chu (1974, Chap. 3).

Two of the more venerable works on multiple scattering are the review article by Milne (1930) and the book by Chandrasekhar (1950). A long-awaited treatise on this subject by van de Hulst (1980) recently made its appearance. Two reports, with many references, edited by Lenoble (1977) and by Fouquart et al. (1980), have been published by the International Association of Meteorology and Atmospheric Physics.

The inverse problem in scattering theory is discussed in the collections of papers edited by Baltes (1978, 1980).

Chapter 2

Electromagnetic Theory

The treatment of absorption and scattering of light by small particles is a problem in electromagnetic theory, a subdivision of which is optics. Various aspects of this theory are found scattered throughout books on electricity and magnetism, optics, and polarization of light. To rely on such existing books as the basis for our development might, however, result in a confusing tangle of conflicting assumptions, notation, and conventions. Also, it is convenient to gather together in one place the mathematical and physical apparatus that underlies succeeding chapters. Thus, we present in this chapter our version of those concepts and equations from electromagnetic theory that are germane to the subject at hand; we shall have need of them throughout the remainder of the book.

2.1 FIELD VECTORS AND THE MAXWELL EQUATIONS

As stated in the introductory chapter, we adopt a macroscopic approach to the problem of determining absorption and scattering of electromagnetic waves by particles. Therefore, the logical point of departure is the Maxwell equations for the macroscopic electromagnetic field at interior points in matter, which in SI units may be written

$$\nabla \cdot \mathbf{D} = \rho_{\mathrm{F}}, \tag{2.1}$$

$$\nabla \times \mathbf{E} + \frac{\partial \mathbf{B}}{\partial t} = 0, \tag{2.2}$$

$$\nabla \cdot \mathbf{B} = 0, \tag{2.3}$$

$$\nabla \times \mathbf{H} = \mathbf{J}_{\mathrm{F}} + \frac{\partial \mathbf{D}}{\partial t}, \tag{2.4}$$

where **E** is the electric field and **B** the magnetic induction. The electric displacement **D** and magnetic field **H** are defined by

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}, \tag{2.5}$$

$$\mathbf{H} = \frac{\mathbf{B}}{\mu_0} - \mathbf{M}, \tag{2.6}$$

where **P** is the electric polarization (average electric dipole moment per unit volume), **M** the magnetization (average magnetic dipole moment per unit volume), ε_0 the permittivity, and μ_0 the permeability of free space. Implicit in (2.5) is the assumption that quadrupole and higher moments are negligible compared with the dipole moment. In free space the polarization and magnetization vanish identically. The charge density ρ_F and current density $\mathbf{J}_F$ are associated with so-called "free" charges. The terms "free" and "bound" are sometimes set in quotation marks, which indicates that they are slightly suspect. Indeed, Purcell (1963, pp. 342–347) has simply but convincingly shown that it is not always possible to unambiguously distinguish between free and bound charges in matter. Nevertheless, we shall assume in time-honored fashion that the ambiguity in the meanings of free and bound leads to no observable consequences in the problems with which we shall be concerned.

Although there appears to be nearly universal agreement about the *microscopic* Maxwell equations, it is not trivial to provide a derivation of the macroscopic equations that will satisfy everyone. The process of so doing was begun by Lorentz some 100 years ago, and new derivations appear regularly in the literature, each of which claims to be more general, freer from ambiguity, and more logically consistent than its predecessors. Without wishing to enter the lists of combatants, we direct the interested reader to Russakoff (1970) and Robinson (1973) for further discussion of the transition from the microscopic to the macroscopic field equations. We accept, therefore, (2.1)–(2.6) as *the* macroscopic field equations without further comment; any attempt to justify this choice rigorously and to define all terms precisely would lead to a volume that would dwarf the present work.

Equations (2.1)–(2.6) are not sufficient in themselves; they must be supplemented with *constitutive relations*, which are assumed to have the form

$$\mathbf{J}_F = \sigma \mathbf{E}, \tag{2.7}$$

$$\mathbf{B} = \mu \mathbf{H}, \tag{2.8}$$

$$\mathbf{P} = \varepsilon_0 \chi \mathbf{E}, \tag{2.9}$$

where σ is the *conductivity*, μ the *permeability*, and χ the electric *susceptibility*. The *phenomenological coefficients* σ, μ, and χ depend on the medium under consideration, but will be assumed to be independent of the fields (the medium is *linear*), independent of position (the medium is *homogeneous*), and independent of direction (the medium is *isotropic*). There are many classes of materials for which these assumptions are *not* valid. Equations (2.7)–(2.9) are not universal laws of nature; they merely describe a particular class of materials which, fortunately, has a large number of members.

We shall not, however, assume that the phenomenological coefficients are independent of *frequency*. To provide some insight into this assertion, we must digress briefly. Consider, for example, the susceptibility. The polarization **P** is the average dipole moment per unit volume of the medium, that is, the vector sum of the dipole moments in a unit volume. An isolated sample of matter (excluding electrets) is unpolarized ($\mathbf{P} = 0$). But when placed in an external field, which we may assume to be time harmonic, it becomes polarized: the electric field induces a net dipole moment. For a linear homogeneous isotropic medium, the relation (2.9) between **P** and **E** indicates that χ is a measure of how easily the material can be polarized; it represents the response of the material to the field **E**. In particular, χ may be interpreted as the amplitude of the response to a unit field. Now it is well known (to those who know it well) that the response of a mechanical system to a periodic driving force is a sensitive function of the frequency. Therefore, by analogy, it is plausible that χ is a function of the frequency. The frequency dependence of χ is discussed in much greater detail in Chapters 9 and 10.

2.2 TIME-HARMONIC FIELDS

The general time-harmonic field **F** has the form

$$\mathbf{F} = \mathbf{A}\cos\omega t + \mathbf{B}\sin\omega t, \tag{2.10}$$

where ω is the *angular frequency*. The *real* vector fields **A** and **B** are independent of time but may depend on position. We note that **F** may be written as the real part of a complex vector: $\mathbf{F} = \mathrm{Re}\{\mathbf{F}_c\}$, where

$$\mathbf{F}_c = \mathbf{C}\exp(-i\omega t), \qquad \mathbf{C} = \mathbf{A} + i\mathbf{B}. \tag{2.11}$$

The vector $\mathbf{F}_c$ is a *complex representation* of the real field **F**. If all our operations on time-harmonic fields are *linear* (e.g., addition, differentiation, integration), it is more convenient to work with the complex representation. The reason this may be done is as follows. Let $\mathfrak{L}$ be any linear operator; we can operate on the field (2.10) by operating on the complex representation (2.11) and then take the real part of the result:

$$\mathfrak{L}\mathbf{F} = \mathfrak{L}\,\mathrm{Re}\{\mathbf{F}_c\} = \mathrm{Re}\{\mathfrak{L}\mathbf{F}_c\}.$$

Note that there is a degree of arbitrariness associated with the complex representation of a real field: **F** could just as easily have been written $\mathbf{F} = \mathrm{Re}\{\mathbf{F}_c^*\}$, where $\mathbf{F}_c^* = \mathbf{C}^*\exp(i\omega t)$ and the asterisk denotes the complex conjugate. Thus, there are two possible choices for the time-dependent factor in a complex representation of a time-harmonic field: $\exp(i\omega t)$ and $\exp(-i\omega t)$. It makes no difference which choice is made: the quantities of physical interest are always real. But once a sign convention has been chosen it must be used consistently in all analysis. We shall take the time-dependent factor to be $\exp(-i\omega t)$; this is the convention found in standard books on optics (Born and Wolf, 1965) and electromagnetic theory (Stratton, 1941; Jackson, 1975) as