1

Introduction

Electromagnetic scattering by an isolated particle or a multi-particle group is a ubiquitous phenomenon central to a wide variety of science and engineering disciplines. Field–matter interactions described by macroscopic electromagnetics typically occur in a natural way. They can affect accompanying physical and chemical processes as well as the very state of the scattering object and often yield an electromagnetic signal that can be measured and analyzed with the purpose of retrieving useful information about the object. Electromagnetic scattering can also be induced artificially and used as an active means of *in situ* or remote diagnostics of certain physical properties of the particle(s). In order to interpret laboratory, field, and remote-sensing measurements of electromagnetic scattering by various single- and multi-particle objects, one needs a deep understanding of this phenomenon, as well as the ability to predict quantitatively its various manifestations as functions of the physical parameters of the objects.

The diversity of sizes, morphologies, and refractive indices of particles encountered in natural and artificial environments is virtually limitless, as illustrated by Fig. 1.1. This factor complicates accurate quantitative modeling of electromagnetic scattering and absorption, even by solitary particles such as those suspended individually in the trap volume of an electrostatic (as shown in Plate 1.1a) or optical levitator. The task of optical modeling of a large group of sparsely distributed particles such as a cloud (see, e.g., Plates 1.1b and 1.1c) is significantly more involved. However, the problem of utmost complexity is to model the scattering and absorption properties of densely packed disperse media such as various biological and technical suspensions (e.g., Plate 1.1d), as well as natural and artificial particulate surfaces (e.g., Plates 1.1e and 1.1f).

The main purpose of this textbook is to coherently present the theory of electromagnetic scattering by solitary particles and particle groups as a self-consistent and self-contained branch of Maxwell's electromagnetics. We will accept the macroscopic Maxwell equations (MMEs) as axioms valid in a well-defined range of applications and will not use any *ad hoc* phenomenological con-

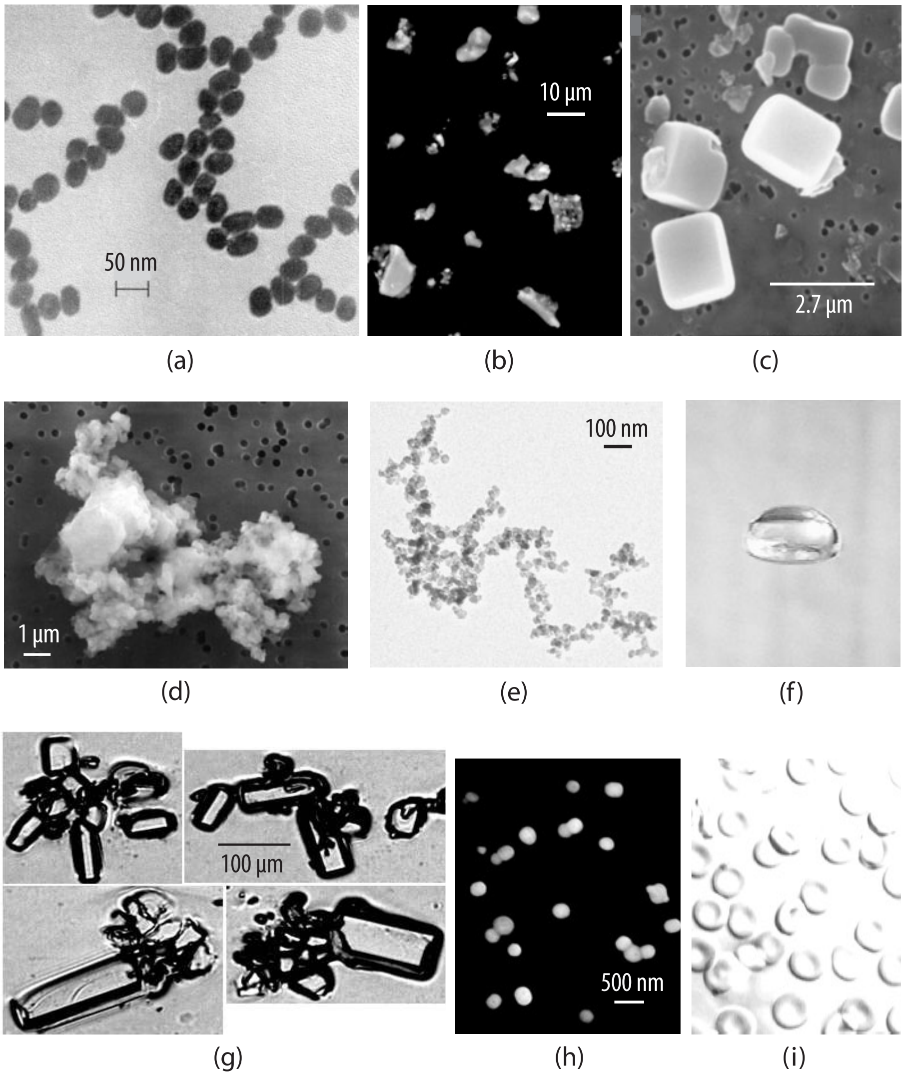

Fig. 1.1. Examples of manmade and natural small particles. **(a)** 40-nm-diameter gold particles (after Khlebtsov *et al.* 1996). **(b)** Sahara-desert soil particles (after Weinzierl *et al.* 2009). **(c)** Dry sea-salt particles (after Chamaillard *et al.* 2003). **(d)** Interplanetary dust particle U2012C11 collected by a NASA U2 aircraft. **(e)** A soot aggregate (after Burr *et al.* 2012). **(f)** A 6-mm-diameter falling raindrop. **(g)** Cirrus cloud crystals (after Arnott *et al.* 1994). **(h)** Submicrometer-sized quasi-spherical ammonium sulphate and dust aerosols (after Weinzierl *et al.* 2009). **(i)** Red blood cells.

cepts and principles not following directly from these equations. This approach is inherently limited in that it ignores specific physical effects described by quantum mechanics and quantum electrodynamics (QED). However, even the intentionally restricted scope of this book will serve to provide accurate quantitative description of many natural and artificially induced electromagnetic scattering effects caused by individual particles and particle groups. Our approach can be called microphysical in that it starts with the MMEs as basic physical laws governing the interaction of electromagnetic radiation with matter and ensures *direct traceability of all derivative results from fundamental physics* not afforded by phenomenological approaches.[1]

There is no doubt that if a specific scattering problem can be addressed by obtaining a direct analytical or numerically exact computer solution[2] of the MMEs, then this would be the preferred course of action. Unfortunately, the range of problems that can be handled analytically is limited. There are several numerically exact techniques for the computer calculation of the electromagnetic field elastically scattered by a finite fixed object composed of one or several particles. These techniques will be reviewed in Chapter 16. However, all of these methodologies have certain practical limitations in terms of the object's morphology and size relative to the wavelength of radiation and cannot be used yet to describe electromagnetic scattering by large multi-particle objects such as atmospheric clouds, particulate surfaces, and particle suspensions. This makes imperative the use of well-characterized approximate solutions that do not require unrealistic computer resources, while being sufficiently accurate for specific applications. One of the objectives of this textbook is to demonstrate that the widely used radiative transfer theory (RTT) as well as the theory of weak localization (WL) of electromagnetic waves in discrete random media are direct asymptotic solutions of the MMEs. Both correspond to the limiting case of a very large number of randomly positioned particles and a very low particle packing density.

1.1 General framework

In contrast to various phenomenological approaches discussed briefly in the following chapters, the microphysical theory of electromagnetic scattering by particles and particle groups pursued in this textbook rests on well-defined assump-

[1] A theory is called phenomenological if it expresses mathematically the results of observed phenomena without paying detailed attention to their fundamental origin and significance. The development of a phenomenological theory is usually based on experience-based heuristic shortcuts lacking rigorous justification. Phenomenological theories are often short-lived and are replaced by fundamental first-principle theories, but some can survive for centuries despite their inherently limited scientific value.

[2] By definition, a numerically exact solution is the outcome of running a direct computer solver of the MMEs that generates numerical results with a guaranteed number of correct decimals. The number of correct decimals may vary depending on the available computer resources and practical accuracy requirements. However, all reported decimals can, in principle, be validated by modifying computer program settings in order to accommodate a more stringent accuracy requirement.

tions intended to formulate the overall problem in strict physical terms. These assumptions are as follows:

1. At each moment in time, the entire scattering object (e.g., a cloud of water droplets or a powder surface) can be represented by a specific spatial configuration of a number $N \geq 1$ of discrete finite particles, as illustrated in Fig. 1.2. The unbounded host medium surrounding the scattering object is homogeneous, isotropic, and nonabsorbing (the general case of an absorbing host medium is discussed by Mishchenko (2007)). Each particle is sufficiently large so that its atomic structure can be ignored and the particle can be characterized by optical constants appropriate to bulk matter. In electromagnetic terms, the presence of a particle means that the optical constants inside the particle volume are different from those of the surrounding host medium. The particles consist of isotropic materials, while their shape and morphology can be arbitrary.

2. Only linear interactions of the electromagnetic field and matter are accounted for. In other words, we will consider only *elastic* electromagnetic scattering. This implies that nonlinear optics effects are excluded by assuming that the optical constants of both the scattering object and the surrounding medium are independent of the electric and magnetic fields. Also excluded are inelastic scattering phenomena, such as Raman and Brillouin scattering and fluorescence, as well as the specific consideration of the small Doppler shift of frequency of the scattered light relative to that of the incident light due to the movement of the particles with respect to the source of illumination. Furthermore, we exclude the phenomenon of thermal emission caused by electron transitions from one energy level to a lower level in macroscopic bodies with absolute temperature different from zero. From the quantum-mechanical standpoint, a macroscopic object is a complex system of molecules with a large number of degrees of freedom. Many different electron transitions produce spectral emission lines so closely spaced that the resulting radiation spectrum becomes effectively continuous and includes

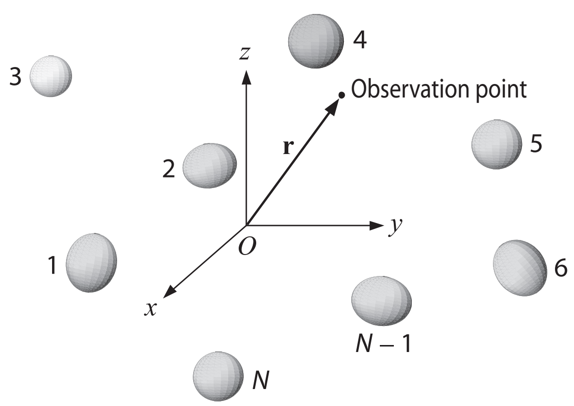

Fig. 1.2. Scattering object in the form of a group of N discrete particles.

emitted energy at all wavelengths. By ignoring thermal emission, we will assume implicitly that the electromagnetic energy at the wavelength in question is much greater than that predicted by the Planck blackbody radiation law. For example, this assumption is usually valid for a short-wave infrared or shorter wavelength in the case of a scattering object at room or lower temperature.

3. Consistent with the restriction of elastic scattering, we will assume that over time intervals much longer than $T_{\rm o} = 2\pi/\omega$, the time dependence of the electric and magnetic fields everywhere in space is harmonic and described, in the complex-field representation, by the simple complex exponential $\exp(-{\rm i}\omega t)$, where ω is the angular frequency, t is time, and ${\rm i} = (-1)^{1/2}$. In other words, we will assume that the complex electric and magnetic fields can be factorized as $\tilde{\boldsymbol{\mathcal{E}}}(\mathbf{r},t) = \exp(-{\rm i}\omega t)\mathbf{E}(\mathbf{r})$ and $\tilde{\boldsymbol{\mathcal{H}}}(\mathbf{r},t) = \exp(-{\rm i}\omega t)\mathbf{H}(\mathbf{r})$, respectively, where $\mathbf{r}$ is the position (radius) vector, while the actual real-valued fields are obtained by taking the real part of the respective complex fields: $\boldsymbol{\mathcal{E}}(\mathbf{r},t) = {\rm Re}\,\tilde{\boldsymbol{\mathcal{E}}}(\mathbf{r},t)$ and $\boldsymbol{\mathcal{H}}(\mathbf{r},t) = {\rm Re}\,\tilde{\boldsymbol{\mathcal{H}}}(\mathbf{r},t)$. The amplitudes $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$ may vary with time implicitly by fluctuating around their respective mean values, but do so much more slowly than the factor $\exp(-{\rm i}\omega t)$. Time-independent amplitudes $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$ correspond to perfectly monochromatic radiation (e.g., a continuous laser beam), whereas the more general case of slowly varying $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$ represents so-called quasi-monochromatic radiation.

4. In addition, we will assume that if the scattering object varies with time then any *significant* (i.e., modifying the solution of the MMEs) changes in particle positions, morphologies, and/or orientations with respect to the laboratory reference frame occur over time intervals $T_{\rm v}$ much longer than the period of time-harmonic oscillations of the electromagnetic field $T_{\rm o}$.

These assumptions imply that over time intervals long compared to $T_{\rm o}$, but short compared to $T_{\rm v}$ and/or to typical periods $T_{\rm f}$ of fluctuations of the amplitudes $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$, all fields can be considered to be perfectly time-harmonic and the object can be considered to be fixed. As a consequence, the instantaneous electromagnetic field everywhere in space can be found by solving the so-called frequency-domain differential MMEs (Stratton 1941; Van Bladel 2007; Rothwell and Cloud 2009) subject to certain boundary conditions. The specific dependence of the optical constants on spatial coordinates and the corresponding boundary conditions at any moment are fully defined by the instantaneous geometrical configuration of the N particles (Fig. 1.2).

Specifically, we will show in Section 4.1 that the frequency-domain monochromatic Maxwell curl equations describing the scattering problem in terms of the time-independent electric and magnetic field amplitudes $\mathbf{E}(\mathbf{r})$ and $\mathbf{H}(\mathbf{r})$ can be written as follows:

$$\left.\begin{aligned} \nabla\times\mathbf{E}(\mathbf{r}) &= {\rm i}\omega\mu_0\,\mathbf{H}(\mathbf{r}) \\ \nabla\times\mathbf{H}(\mathbf{r}) &= -{\rm i}\omega\epsilon_1\,\mathbf{E}(\mathbf{r}) \end{aligned}\right\}\quad \mathbf{r}\in V_{\rm EXT}, \tag{1.1}$$

$$\left.\begin{aligned}\nabla\times\mathbf{E}(\mathbf{r}) &= \mathrm{i}\omega\mu_0\,\mathbf{H}(\mathbf{r})\\ \nabla\times\mathbf{H}(\mathbf{r}) &= -\mathrm{i}\omega\varepsilon_2(\mathbf{r},\omega)\,\mathbf{E}(\mathbf{r})\end{aligned}\right\}\quad \mathbf{r}\in V_{\mathrm{INT}}. \tag{1.2}$$

In these equations, V_{INT} is the cumulative "interior" volume occupied by the scattering object; V_{EXT} is the infinite exterior region such that $V_{\mathrm{INT}} \cup V_{\mathrm{EXT}} = \Re^3$, where $\Re^3$ denotes the entire three-dimensional space; the host medium and the scattering object are assumed to be nonmagnetic; μ_0 is the permeability of a vacuum; ϵ_1 is the real-valued electric permittivity of the host medium; and $\varepsilon_2(\mathbf{r},\omega)$ is the (potentially coordinate-dependent) complex permittivity of the scattering object.

Equations (1.1) and (1.2) supplemented by the so-called radiation condition at infinity, as well as by the standard boundary conditions for the electric and magnetic fields defined by the specific spatial distribution of the refractive index have a solution, this solution being unique (Müller 1969). This fundamental fact makes the MMEs a self-sufficient basis of the electromagnetic scattering theory. In other words, the rest of this book will be an outline of analytical and numerical solutions of Eqs. (1.1) and (1.2) (or their integral-equation counterparts) as applied to scattering objects of varying complexity.

1.2 Electromagnetic scattering

In the absence of the scattering object, $\varepsilon_2(\mathbf{r},\omega) \equiv \epsilon_1$. This means that instead of Eqs. (1.1) and (1.2), we have:

$$\left.\begin{aligned}\nabla\times\mathbf{E}(\mathbf{r}) &= \mathrm{i}\omega\mu_0\,\mathbf{H}(\mathbf{r})\\ \nabla\times\mathbf{H}(\mathbf{r}) &= -\mathrm{i}\omega\epsilon_1\,\mathbf{E}(\mathbf{r})\end{aligned}\right\}\quad \mathbf{r}\in \Re^3. \tag{1.3}$$

Obviously, the solution of Eq. (1.3) must differ from that of the system of equations (1.1) and (1.2). It is this *modification* of the electromagnetic field resulting from the presence of the object that is called *electromagnetic scattering*.

We thus see that the specific way to define electromagnetic scattering is to solve the MMEs twice. The first solution, in terms of the respective pair of the electric and magnetic fields $\{\mathbf{E}_1(\mathbf{r}), \mathbf{H}_1(\mathbf{r})\}$, corresponds to the situation with no scattering object (Eq. (1.3)), whereas the second solution, $\{\mathbf{E}_2(\mathbf{r}), \mathbf{H}_2(\mathbf{r})\}$, corresponds to the situation with a scattering object present (Eqs. (1.1) and (1.2)).

The second solution is usually sought in the form

$$\mathbf{E}_2(\mathbf{r}) = \mathbf{E}_1(\mathbf{r}) + \mathbf{E}_3(\mathbf{r}), \tag{1.4}$$

$$\mathbf{H}_2(\mathbf{r}) = \mathbf{H}_1(\mathbf{r}) + \mathbf{H}_3(\mathbf{r}), \tag{1.5}$$

where the vector fields $\mathbf{E}_3(\mathbf{r})$ and $\mathbf{H}_3(\mathbf{r})$ are required to satisfy the radiation condition at infinity by decaying as the inverse distance from the object. The electromagnetic field in the absence of the object is traditionally called the "incident field" (superscript "inc"), whereas the difference between the electromagnetic field in the presence of the object and the field that would exist in the absence of the object is traditionally called the "scattered field" (superscript "sca"):

$$\mathbf{E}^{\rm sca}(\mathbf{r}) \equiv \mathbf{E}_3(\mathbf{r}) = \mathbf{E}_2(\mathbf{r}) - \mathbf{E}_1(\mathbf{r}) = \mathbf{E}_2(\mathbf{r}) - \mathbf{E}^{\rm inc}(\mathbf{r}), \tag{1.6}$$

$$\mathbf{H}^{\rm sca}(\mathbf{r}) \equiv \mathbf{H}_3(\mathbf{r}) = \mathbf{H}_2(\mathbf{r}) - \mathbf{H}_1(\mathbf{r}) = \mathbf{H}_2(\mathbf{r}) - \mathbf{H}^{\rm inc}(\mathbf{r}). \tag{1.7}$$

The above discussion makes it clear that although electromagnetic scattering can be said to be a physical *phenomenon* (amounting to the *fact* that the electromagnetic fields computed in the presence and in the absence of an object are different), it is not a solitary physical process. Importantly, Eqs. (1.6) and (1.7) demonstrate again that the electromagnetic field in the presence of the object is intentionally expressed as the sum of the incident and scattered fields:

$$\mathbf{E}(\mathbf{r}) \equiv \mathbf{E}_2(\mathbf{r}) = \mathbf{E}^{\rm inc}(\mathbf{r}) + \mathbf{E}^{\rm sca}(\mathbf{r}), \tag{1.8}$$

$$\mathbf{H}(\mathbf{r}) \equiv \mathbf{H}_2(\mathbf{r}) = \mathbf{H}^{\rm inc}(\mathbf{r}) + \mathbf{H}^{\rm sca}(\mathbf{r}), \tag{1.9}$$

where $\mathbf{E}^{\rm inc}(\mathbf{r})$ and $\mathbf{H}^{\rm inc}(\mathbf{r})$ are obtained by solving the MMEs in the absence of the object. This makes both the incident and the scattered field, as they appear in the solution of Eqs. (1.1) and (1.2) in the form of Eqs. (1.8) and (1.9), purely mathematical entities rather than actual physical fields. The only actual field is the *total* electromagnetic field, either in the presence or in the absence of the object.

This basic point is illustrated in Fig. 1.3. Indeed, a fundamental solution of the MMEs in an infinite nonabsorbing homogeneous space is a time-harmonic plane electromagnetic wave given, in the complex-field representation, by

$$\left.\begin{aligned} \tilde{\boldsymbol{\mathcal{E}}}^{\rm inc}(\mathbf{r},t) &= \mathbf{E}_0^{\rm inc}\exp(\mathrm{i}\mathbf{k}^{\rm inc}\cdot\mathbf{r} - \mathrm{i}\omega t) \\ \tilde{\boldsymbol{\mathcal{H}}}^{\rm inc}(\mathbf{r},t) &= \mathbf{H}_0^{\rm inc}\exp(\mathrm{i}\mathbf{k}^{\rm inc}\cdot\mathbf{r} - \mathrm{i}\omega t) \end{aligned}\right\} \quad \mathbf{r}\in\Re^3 \tag{1.10}$$

with constant complex amplitudes $\mathbf{E}_0^{\rm inc}$ and $\mathbf{H}_0^{\rm inc}$, where $\mathbf{k}^{\rm inc}$ is a real-valued so-called wave vector. In fact, this solution embodies the concept of a perfectly monochromatic parallel beam of light of infinite lateral extent propagating in the direction of the wave vector. The time-independent part of this solution is visualized in Fig. 1.3a. Placing a spherical particle at the origin of the coordinate system, as shown in Fig. 1.3c, yields a new time-harmonic solution of the MMEs. The time-independent part of this second solution, both inside and outside the particle volume, is visualized in Fig. 1.3b. Subtracting the field visualized in Fig. 1.3a from that depicted in Fig. 1.3b yields the scattered field shown in Fig. 1.3c. It is clear that the fields in Figs. 1.3a and 1.3b are actual physical fields found by solving the MMEs, whereas the field in Fig. 1.3c is a purely mathematical construction.

A fundamental corollary of the above discussion is that, irrespective of the morphological complexity of the scattering object, the latter always remains a single, unified scatterer. Although the human eye may classify a scattering object

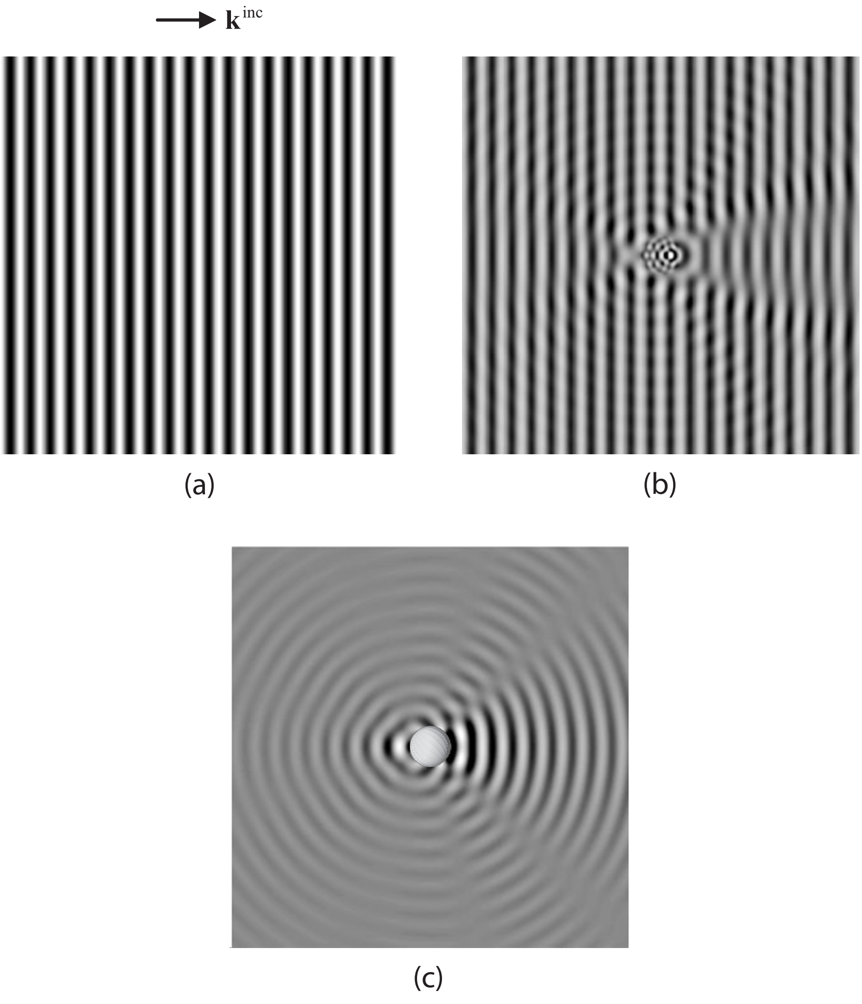

Fig. 1.3. **(a)** The real part of the vertical (i.e., perpendicular to the paper) component of the electric field vector of a plane electromagnetic wave propagating in the direction of the wave vector $\mathbf{k}^{\text{inc}}$. The infinite host medium is homogeneous, isotropic, and nonabsorbing. The wave is fully polarized in the vertical direction so that the horizontal component of the electric field vector is equal to zero. **(b)** The real part of the vertical component of the total electric field in the presence of a small homogeneous spherical particle located in the center of the diagram as to be shown in panel (c). The relative refractive index of the particle is $m = (\varepsilon_2/\epsilon_1)^{1/2} = 2.8$, while its radius a is such that the dimensionless size parameter $k_1 a = \omega(\epsilon_1 \mu_0)^{1/2} a$ is equal to 2π. **(c)** The real part of the vertical component of the difference between the fields visualized in panels (b) and (a), respectively. The gray scale is individually adjusted to maximally reveal the specific details in each diagram. (After Mishchenko and Travis (2008); images courtesy of J.-C. Auger.)

(e.g., a cloud of water droplets or ice crystals) as a "collection of discrete particles," the incident field perceives the entire object at any moment in time as one scatterer in the form of a specific instantaneous spatial distribution of the complex permittivity $\varepsilon_2(\mathbf{r}, \omega)$. This means that any of the "multi-particle groups" shown in Plates 1.1b–1.1f is as much a single-scattering object as the solitary particle shown in Plate 1.1a.

This corollary implies that the widely used term "multiple scattering" does not refer to an actual physical phenomenon. We will see in later chapters that it has only a purely mathematical meaning.

1.3 Further remarks

Throughout most of this book, we will consider the incident field in the form of a perfectly monochromatic plane wave given by Eq. (1.10). More generally, the incident field can be a superposition of monochromatic or quasi-monochromatic plane waves. A quasi-monochromatic plane wave is given by

$$\left.\begin{aligned}\tilde{\boldsymbol{\mathcal{E}}}^{\text{inc}}(\mathbf{r},t) &= \mathbf{E}_0^{\text{inc}}(t)\exp(\mathrm{i}\mathbf{k}^{\text{inc}}\cdot\mathbf{r}-\mathrm{i}\omega t)\\ \tilde{\boldsymbol{\mathcal{H}}}^{\text{inc}}(\mathbf{r},t) &= \mathbf{H}_0^{\text{inc}}(t)\exp(\mathrm{i}\mathbf{k}^{\text{inc}}\cdot\mathbf{r}-\mathrm{i}\omega t)\end{aligned}\right\}\quad \mathbf{r}\in\Re^3, \tag{1.11}$$

where fluctuations in time of the complex amplitudes of the electric and magnetic fields, $\mathbf{E}_0^{\text{inc}}(t)$ and $\mathbf{H}_0^{\text{inc}}(t)$, around their respective mean values are assumed to occur much more slowly than the time-harmonic oscillations of the factor $\exp(-\mathrm{i}\omega t)$. We will not consider in detail other types of incident field such as a focused laser beam of finite lateral extent (e.g., Gouesbet and Gréhan 2011) or an ultra-short pulse. However, as will be discussed in the following chapters, this restriction narrows the scope of the book not nearly as much as it may appear at first glance.

It is worth emphasizing again that macroscopic Maxwell's electromagnetics ignores the discreteness of matter forming the scattering object, operates with continuous sources of fields, and captures only linear field–matter interactions. Therefore, its predictions can fall short in cases where quantum effects are essential. Even so, the quantum theory can often be used to determine the macroscopic electromagnetic properties of bodies consisting of very large numbers of atoms (Akhiezer and Peletminskii 1981; Lukš and Peřinová 2009). It turns out that this approach works well when the external electromagnetic field is sufficiently weak and the particle size exceeds ~50 Å (Huffman 1988). This result is very important since it: (i) implies a rather wide range of applicability of the MMEs and (ii) allows one to clarify the definition of a "macroscopic particle" as the subject of study in this book in terms of the smallest allowable particle size.

We can thus conclude that the use of macroscopic Maxwell's electromagnetics as the point of departure is founded on the well-established fact that this theory follows directly from more fundamental physical theories as a consequence of well-characterized and verifiable approximations. This fact allows us to adopt the equations of classical macroscopic electromagnetics essentially as basic axioms valid in a wide and well-defined range of practical situations. The reader will see that this approach can be used to develop a mostly self-contained and self-consistent theory in which the need to invoke alternative physical concepts and laws is largely obviated.

1.4 Energy-budget and optical-characterization problems

A clear understanding of the type of field–matter interactions captured by the MMEs helps identify the extent of their applicability to solving specific practical problems. Let us consider, for example, an imaginary liquid-water cloud illuminated by a parallel quasi-monochromatic beam of light with infinite lateral extent, as shown schematically in Fig. 1.4. Suppose that we need to evaluate the energy budget of a macroscopic volume element ΔV bounded by the closed surface ΔS. According to the Poynting theorem discussed specifically in Section 2.4, the net

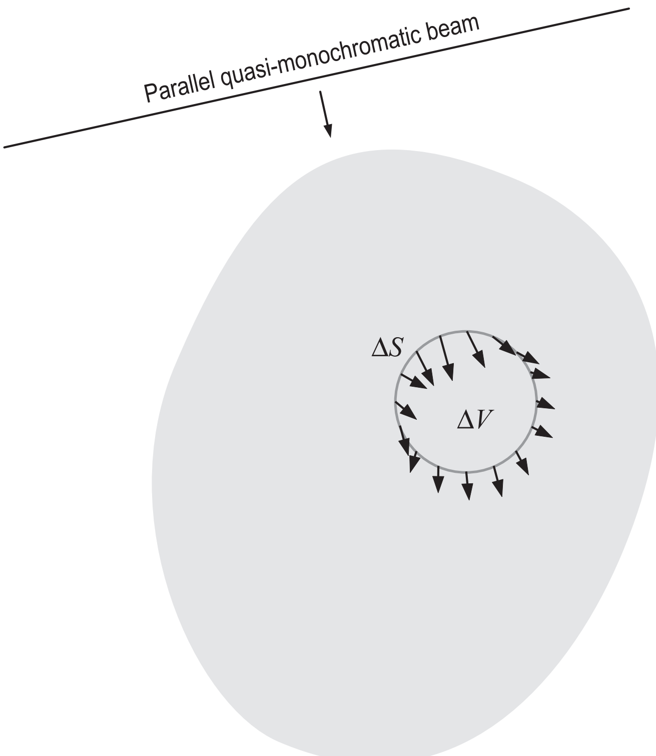

Fig. 1.4. Time-averaged energy budget of a volume element ΔV of a cloud bounded by the closed surface ΔS. The arrows represent the distribution of $\langle\langle \boldsymbol{\mathcal{S}}(\mathbf{r},t)\rangle\rangle$ over the boundary ΔS.