Gustav Mie and the fundamental concept of electromagnetic scattering by particles: A perspective

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Review

Gustav Mie and the fundamental concept of electromagnetic scattering by particles: A perspective

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A B S T R A C T

This tutorial review provides a general discussion of the fundamental concept of electromagnetic scattering by particles and particle groups and dispels certain widespread yet profoundly confusing misconceptions.

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1. Introduction

Since the seminal paper by Gustav Mie [1], light (or, more generally, electromagnetic) scattering by macroscopic particles has been the subject of innumerable publications (e.g., [2–43] and references therein). However, the very notion of "scattering" has rarely been defined explicitly and is usually assumed to be intuitively obvious to the reader. This is true of journal papers as well as books (see, e.g., [3, Section 1.1]). There are, of course, exceptions. For example, Davis and

Knyazikhin [44, p. 170] assume that "upon collision with an atmospheric particle, a photon can be either absorbed or scattered." They then go on to define scattering as, "a random choice of new direction of propagation for the photon" (p. 178). The gist of this representation of scattering as a "collision" of a light corpuscle with a cloud droplet followed by the corpuscle changing the direction of flight (Fig. 1a) is succinctly summarized by Bohren and Clothiaux [45, p. 295]: "an incident photon becomes a scattered photon." This neo-Newtonian concept of light scattering by a macroscopic particle appears to be appealing and is rather common. However, as we will demonstrate below, it falls apart upon a closer look at what the authors mean by a "photon".

Another attempt at an explicit definition of scattering is made in the fundamental encyclopedia of scattering by Pike and Sabatier [46, p. xi]: "Suppose we have a complete knowledge of an incident field of waves or particles as they are emitted
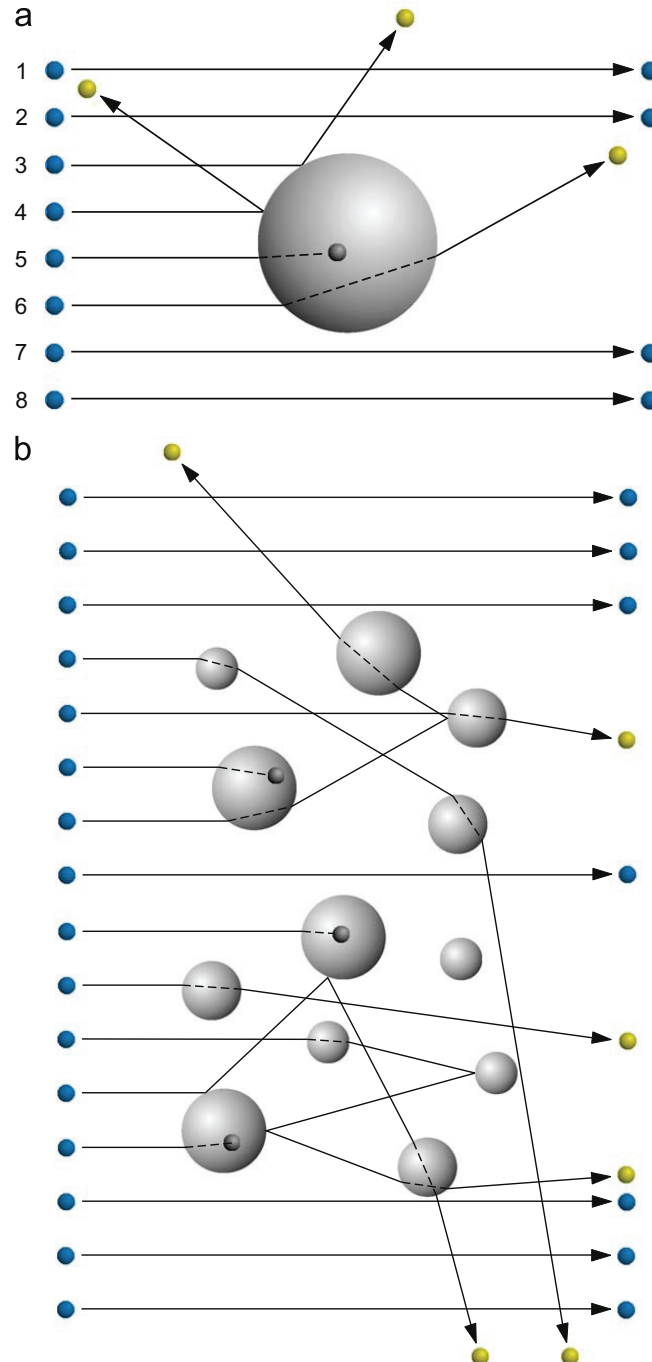

Fig. 1. (a) Interaction of a "photonic beam" with a macroscopic particle. Photons 1, 2, 7, and 8 "pass" without scattering. Photons 3, 4, and 6 are scattered by the particle. Photon 5 is absorbed inside the particle. (b) Multiple scattering of a "photonic beam" by a group of particles.

from a source into a more or less penetrable obstacle or set of obstacles collectively called a target. After they interact with the target, the incident field will be turned into outgoing waves or particles called the emergent field. We call this process scattering." This definition also appears to be consistent with the traditional intuitive perception of scattering. Still, as we shall see later, there are fundamental reasons to question the general relevance of the words "after", "interaction", "into", and "process" which make this definition inapplicable to electromagnetic scattering by particles. Similarly, Bohren and Huffman [8, p. 3–4] assert erroneously that scattering = excitation by the incident wave+reradiation. Mishchenko et al. [14, p. 3] repeat this false assertion.

The disciplines of multiple scattering and radiative transfer can be even more confusing in their reliance on the intuitive perception of successive scattering events caused by a sequence of particles. For example, van de Hulst [47, p. ix] defines the subject of the radiative transfer theory as "the play of radiation by repeated scattering in a cloud layer or any other slab of particles." Thomas and Stamnes [48] discussed probabilistic aspects of radiative transfer in terms of a "photon" executing a multiple scattering trajectory, depending upon the random nature of the angular scattering process (Fig. 1b). Petty [49] also gives specific examples of random paths of "photons" going through various sequences of scattering centers. We will see later, however, that multiple scattering is a purely mathematical construction rather than a real physical process.

The main objective of this tutorial paper is to clarify the definitions of single and multiple scattering of light by macroscopic particles and particle groups. Following Mie [1], we will operate in the framework of macroscopic Maxwell's electromagnetics, thereby obviating the need for the (often confusing) notion of photons as well as for the microscopic treatment in terms of elementary electric charges.

2. Frequency-domain scattering by a particle

In modern physical terms, the Mie theory as well as its various generalizations [1,3,4,8,12,14,17,18,20,24] belong in the realm of so-called frequency-domain macroscopic electromagnetics [2,50]. This means that all fields and sources of fields are assumed to vary in time harmonically (i.e., are proportional to a common factor $\exp(-i\omega t)$, where ω is the angular frequency, t is time, and $i = (-1)^{1/2}$). Furthermore, the scattering object is defined as a finite volume with a refractive index different from that of the surrounding infinite homogeneous medium. The fundamental concept of electromagnetic scattering used by Mie can be illustrated as follows [51]: A basic solution of the macroscopic Maxwell equations is a plane electromagnetic wave propagating in an infinite nonabsorbing medium without a change in its intensity or polarization state (Fig. 2a). However, in the presence of a particle the electromagnetic field differs from that corresponding to the unbounded homogeneous space (Fig. 2b). The difference between the total field in the presence of the particle and the original field that would exist in the absence of the particle might be thought of as the field scattered by the particle (Fig. 2c). In other words, the total field in the presence of the particle is represented as the sum of the respective incident (original) and scattered fields:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^{\text{inc}}(\mathbf{r}) + \mathbf{E}^{\text{sca}}(\mathbf{r}), \tag{1}$$

$$\mathbf{H}(\mathbf{r}) = \mathbf{H}^{\text{inc}}(\mathbf{r}) + \mathbf{H}^{\text{sca}}(\mathbf{r}), \tag{2}$$

where $\mathbf{E}$ is the electric field, $\mathbf{H}$ is the magnetic field, $\mathbf{r}$ is the position vector, and the common factor $\exp(-i\omega t)$ is omitted. Thus, it is the *modification* of the *total* electromagnetic field caused by the presence of the particle that is called *electromagnetic scattering*.

It should be clearly understood that the separation of the total field in the presence of the particle into the incident and scattered components has a purely mathematical character. In other words, the fields on the right-hand sides of Eqs. (1) and (2) are not real physical fields. Thus, although frequency-domain electromagnetic scattering by a macroscopic particle can be said to be a physical *phenomenon* (amounting to the *fact* that the total field computed in the presence of a particle is different from that computed in the absence of the particle), it is not a solitary physical process.

This explains why any measurement of electromagnetic scattering is always reduced to the measurement of certain optical observables first in the absence of the particle and then in the presence of the particle, as illustrated in Fig. 3. The differences between the readings of detectors of electromagnetic energy quantify the scattering and absorption properties of the particle and can often be interpreted in order to infer the particle microphysical properties. The first measurement stage can sometimes be implicit (e.g., it is often bypassed by assuming that the reading of detector 2 in the absence of the scattering object is zero) or is called "detector calibration", but this does not change the two-stage character of any scattering measurement [52–55]. Typically, detector 1 facing the incident wave would be used to measure the particle extinction matrix, while detector 2 would be used to measure the angular dependence of the phase (or scattering) matrix provided that both detectors are located in the far-field zone of the particle [3,14].

Of course, one can generalize the concept of electromagnetic scattering by considering an incident field $\mathbf{E}^{\text{inc}}(\mathbf{r})$ other than a plane wave. This can be done in a straightforward way using the so-called volume integral equation (VIE) [14,56] which follows directly from the frequency-domain macroscopic Maxwell equations and incorporates the requisite boundary conditions at the particle surface as well as the so-called radiation conditions at infinity [57,58]. The
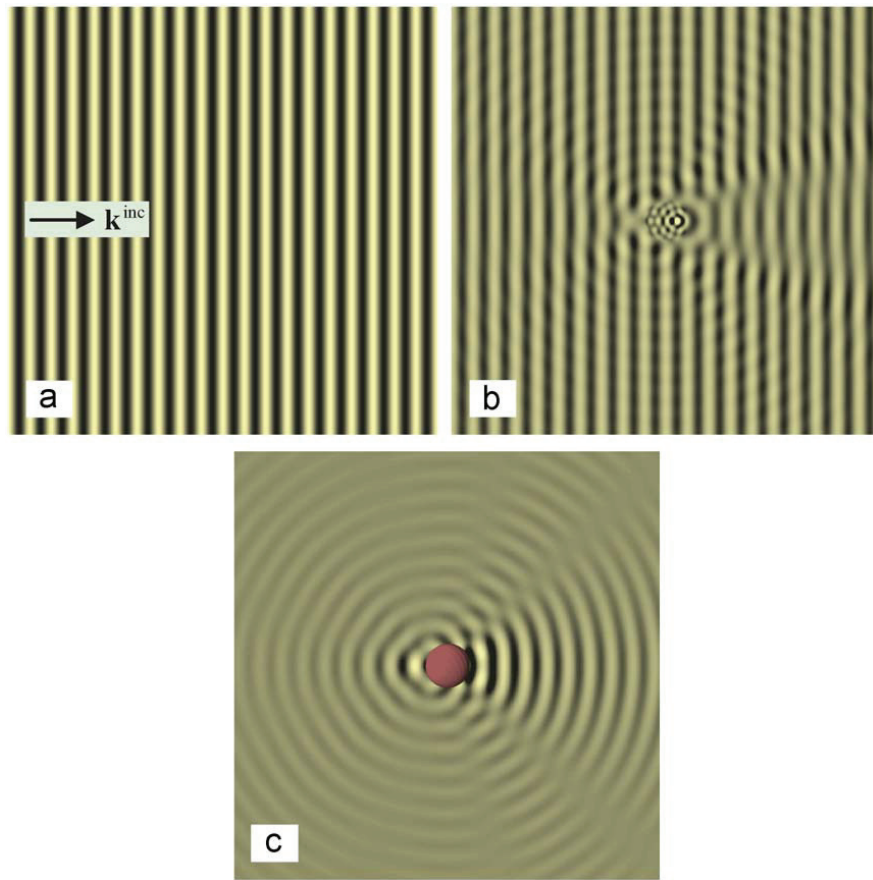
Fig. 2. (a) The real part of the vertical (i.e., perpendicular to the paper) component of the electric field vector of a plane electromagnetic wave propagating in the direction of the wave vector $\mathbf{k}^{\text{inc}}$. The wave is fully polarized in the vertical direction so that the horizontal component 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 shown in panel (c). The relative refractive index of the particle is 2.8, while its radius a is such that the size parameter k_1a is equal to 2π, where k_1 is the wave number in the host medium. (c) The real part of the vertical component of the difference between the fields visualized in panels (b) and (a). The color scale was individually adjusted to maximally reveal the specific details in each diagram.

latter are necessary in order to ensure the uniqueness of solution of the Maxwell equations. The VIE for the electric field reads

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^{\text{inc}}(\mathbf{r}) + k_1^2 \int_{V_{\text{INT}}} \mathrm{d}\mathbf{r}' \, \overset{\leftrightarrow}{G}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{E}(\mathbf{r}')[m^2(\mathbf{r}') - 1]$$

$$= \mathbf{E}^{\text{inc}}(\mathbf{r}) + k_1^2 \left(\overset{\leftrightarrow}{I} + \frac{1}{k_1^2} \nabla \otimes \nabla \right) \cdot \int_{V_{\text{INT}}} \mathrm{d}\mathbf{r}' \mathbf{E}(\mathbf{r}') \frac{\exp(ik_1|\mathbf{r} - \mathbf{r}'|)}{4\pi|\mathbf{r} - \mathbf{r}'|} [m^2(\mathbf{r}') - 1], \quad \mathbf{r} \in \mathfrak{R}^3, \tag{3}$$

where $m(\mathbf{r}')$ is the refractive index of the particle interior relative to that of the host exterior medium, k_1 is the wave number in the host medium, $\overset{\leftrightarrow}{G}(\mathbf{r}, \mathbf{r}')$ is the free space dyadic Green's function, $\overset{\leftrightarrow}{I}$ is the identity dyadic, $\otimes$ is the dyadic product sign, V_{INT} is the interior volume of the particle, and $\mathfrak{R}^3$ is the entire three-dimensional space. The VIE expresses the total field everywhere in space in terms of the total field inside the scattering object. If the scattering object is absent then $m(\mathbf{r}') \equiv 1$, and the total field is identically equal to the incident field. Otherwise the total field contains a scattering component given by the second term on the right-hand side of Eq. (3).

As we have already mentioned, it is often said that the incident field is transformed *into* the scattered field upon its interaction with the object (e.g., [46]). This assertion usually serves to emphasize an alleged causality of the "scattering process": the incident field is presented as the cause of the scattered field. This is also the gist of the definition of scattering given by Bohren and Huffman [8, p. 3–4]: scattering = excitation by the incident wave+reradiation. They further assert that in addition to reradiating electromagnetic energy, the object may transform part of the incident electromagnetic energy into other forms of energy (e.g., thermal energy) owing to absorption.

However, the very representation of the total field in the presence of the object as the superposition of the incident and scattered fields according to Eqs. (1) and (2) implies that the incident field and hence its energy *remain unchanged* rather than are transformed into the scattered field and absorbed energy. Furthermore, as we have seen, the incident field in Eqs. (1) and (2) is a purely *mathematical* quantity and cannot *physically* interact with the particle and "excite" it. It also
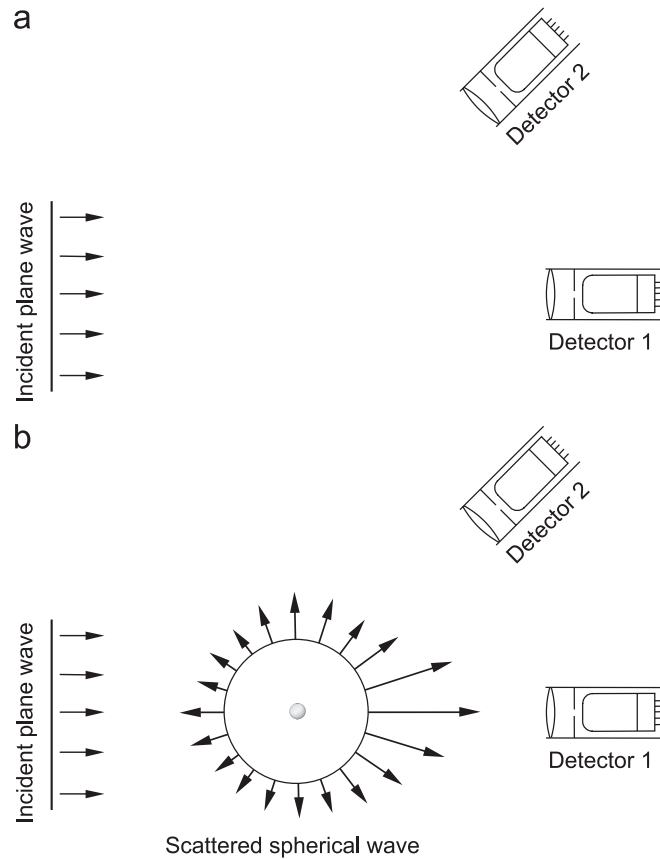
Fig. 3. The readings of detectors of electromagnetic energy in the presence of a particle (diagram (b)) differ from those in the absence of the particle (diagram (a)).

cannot "cause" the scattered field [59]. Even if one wants to invoke the concept of "excitation" then Eq. (3) clearly shows that the particle is excited by the total field rather than by the incident field. In fact, it is meaningless to talk about energy contained in the physically non-existent incident field; it is the energy of the total field that gets reduced by absorption, not that of the incident field.

3. Time-domain scattering by a particle

The fundamental concept of electromagnetic scattering discussed above remains valid in the case of transient rather than time-harmonic fields (i.e., in the framework of time-domain macroscopic electromagnetics [50,60,61]) and is embodied by the same measurement configuration (Fig. 3). As before, in order to facilitate the theoretical interpretation of transient measurements, the time-domain macroscopic Maxwell equations are solved twice. The first solution, $\{\mathbf{E}_1, \mathbf{H}_1\}$, corresponds to the situation with no scattering object, whereas the second solution, $\{\mathbf{E}_2, \mathbf{H}_2\}$, corresponds to the situation with a scattering object present and is intentionally sought in the form $\{\mathbf{E}_2 = \mathbf{E}_1 + \mathbf{E}_3, \ \mathbf{H}_2 = \mathbf{H}_1 + \mathbf{H}_3\}$, where the fields $\mathbf{E}_3$ and $\mathbf{H}_3$ are required to satisfy the radiation conditions at infinity. It is understood again that the difference between the solutions $\{\mathbf{E}_1, \mathbf{H}_1\}$ and $\{\mathbf{E}_2, \mathbf{H}_2\}$ is caused by the differences in the corresponding initial and boundary conditions.

As before, the fields $\mathbf{E}_3$ and $\mathbf{H}_3$ are obtained by means of a purely mathematical subtraction of electric and magnetic fields corresponding to two quite different physical situations: $\mathbf{E}_3 = \mathbf{E}_2 - \mathbf{E}_1$ and $\mathbf{H}_3 = \mathbf{H}_2 - \mathbf{H}_1$. Calling $\{\mathbf{E}_1, \mathbf{H}_1\}$ the "incident field" and $\{\mathbf{E}_3, \mathbf{H}_3\}$ the "scattered field" is a matter of convention and does not mean that the scattered field actually exists and is caused by the incident field. Indeed, there can be no temporal causal relation between two separate solutions of the Maxwell equations $\{\mathbf{E}_1, \mathbf{H}_1\}$ and $\{\mathbf{E}_2, \mathbf{H}_2\}$. Hence there can be no temporal causal relation between $\{\mathbf{E}_1, \mathbf{H}_1\}$ and $\{\mathbf{E}_3, \mathbf{H}_3\}$.

Time-domain electromagnetic scattering is often pictured at an intuitive level as a solitary physical process unfolding in time. This involves a preceding incoming wave, an interaction of this wave with the particle, and a subsequent outgoing scattered wave, thereby allowing for the description of the scattering process by two separate, causally related fields. However, this picture is profoundly misleading because it suggests that the "incident" and "scattered" fields are what we would observe in the physical world. In reality, however, there is only one observable electromagnetic field, viz., the total field. The "incident" and "scattered" fields in the presence of the object are only formally defined mathematical quantities with no direct physical counterparts.

Finally, in complete analogy with the frequency-domain case, the scattering object is "excited" by the total field rather than by the incident field (see Eqs. (4.35) and (4.36) of [60]).

4. Multiple scattering

The mathematical origin of the "multiple scattering" terminology in the radiative transfer theory can be traced back to the frequency-domain so-called Foldy–Lax equations applied to a scattering object composed of N geometrically non-overlapping particles. These equations follow directly from the VIE [17,62] and can be used to derive the following "order-of-scattering expansion" of the total electric field at an observation point [63,64]:

$$E = E^{\mathrm{inc}} + E^{\mathrm{sca}}, \tag{4}$$

$$E^{\mathrm{sca}} = \sum_{i=1}^{N} \hat{G}\hat{T}_i E^{\mathrm{inc}} + \sum_{\substack{i=1 \\ j(\neq i)=1}}^{N} \hat{G}\hat{T}_i \hat{G}\hat{T}_j E^{\mathrm{inc}} + \sum_{\substack{i=1 \\ j(\neq i)=1 \\ l(\neq j)=1}}^{N} \hat{G}\hat{T}_i \hat{G}\hat{T}_j \hat{G}\hat{T}_l E^{\mathrm{inc}} + \cdots, \tag{5}$$

where a compact operator notation is used. Specifically, $\hat{G}$ represents the free space dyadic Green's function, $\hat{T}_i$ represents the so-called dyadic transition operator $\overleftrightarrow{T}_i(\mathbf{r}', \mathbf{r}'')$ of particle i [62,63], and

$$\hat{G}\hat{T}_j E = \int_{V_j} \mathrm{d}\mathbf{r}' \, \overleftrightarrow{G}(\mathbf{r}, \mathbf{r}') \cdot \int_{V_j} \mathrm{d}\mathbf{r}'' \, \overleftrightarrow{T}_j(\mathbf{r}', \mathbf{r}'') \cdot \mathbf{E}(\mathbf{r}''), \tag{6}$$

where V_j is the interior volume of particle j. The dyadic transition operators are independent of each other, and each of them can be interpreted as a complete individual electromagnetic identifier of the corresponding particle. It is, therefore, tempting to interpret $\hat{G}\hat{T}_i E^{\mathrm{inc}}$ as the partial scattered field at the observation point generated by particle i in response to the "excitation" by the incident field only, $\hat{G}\hat{T}_i \hat{G}\hat{T}_j E^{\mathrm{inc}}$ as the partial field generated by the same particle in response to the "excitation" caused by particle j in response to the "excitation" by the incident field, etc. The first term on the right-hand side of Eq. (4) then represents the unscattered (i.e., incident) field.

The use of the "multiple scattering" terminology may be a convenient and a compact way of illustrating the numerous consequences of the Foldy–Lax equations, in particular the microphysical theories of radiative transfer and coherent backscattering [63,64]. However, it follows from the Flody–Lax equations that all mutual particle–particle "excitations" occur simultaneously and are not temporally discrete and ordered events. The purely mathematical character of the multiple scattering interpretation of Eqs. (4) and (5) becomes especially obvious upon realizing that these equations are quite general and can be applied not only to a multi-particle group but also to a single body wherein the latter is subdivided artificially into N non-overlapping adjacent geometrical regions V_i.

Thus, the concept of multiple scattering is as much a purely mathematical abstraction as the incident and scattered fields. This conclusion applies equally to transient electromagnetic scattering by a multi-particle configuration.

5. Validity of Mie's scattering concept

The fundamental concept of electromagnetic scattering by a particle pursued by Gustav Mie has been verified, explicitly or implicitly, in countless publications (see, e.g., [1–43] and references therein). Perhaps the most spectacular validation is the ability of the Mie theory to explain, both qualitatively and quantitatively, the magnificent atmospheric optical displays such as rainbows, fogbows, and the glory caused by spherical water droplets (see Fig. 4 and [65–68]). A perfect quantitative way to validate the Mie theory with extreme precision is to measure and calculate various manifestations of so-called morphology-dependent resonances (MDRs) for homogeneous as well as layered spherical particles [69–74]. Fig. 5a gives an impressive example of the ability of the Mie theory to reproduce observed side-scattering intensity spectra for a gradually evaporating glycerol droplet. In fact, the measurement and analysis of super-narrow MDRs turns out to be the most accurate tool for the determination of particle size, refractive index, internal structure, and nonsphericity [70–75]. Equally definitive is the validation of the electromagnetic scattering concept in general and the scale invariance rule [76] in particular by using fully controlled laboratory measurements at microwave frequencies [54,55,77].

The recently developed microphysical approach to radiative transfer (see [63,64] and references therein) has demonstrated that by applying the fundamental electromagnetic scattering concept to a large, fully ergodic random group of sparsely distributed particles, one can derive the vector radiative transfer equation (RTE) without invoking any additional phenomenological conceptions (e.g., "elementary volume elements" and "photons"). This means, in particular, that although the RTE has the formal structure of a kinetic equation typically associated with a particle transport process, it belongs in the realm of electromagnetic wave scattering.

One of the classical applications of the RTE in remote sensing is the analysis of ground-based multi-spectral polarimetric observations of Venus covering a wide scattering-angle range from almost 0° to essentially 180°. By comparing the results of polarimetric measurements with numerically accurate computer solutions of the RTE, Hansen and Hovenier [78] were able to determine the size distribution, shape, and refractive index of cloud particles in the Venus atmosphere with extreme precision (see Fig. 5b). The retrieved spectral behavior of the refractive index led to an unequivocal identification of the

Fig. 4. The upper panel shows a rainbow photographed from a helicopter above the Big Island of Hawaii. The bottom panel shows a glory, a Brocken Spectre, and a fogbow photographed from San Francisco's Golden Gate Bridge. Photographs courtesy of Lyudmila Zinkova.
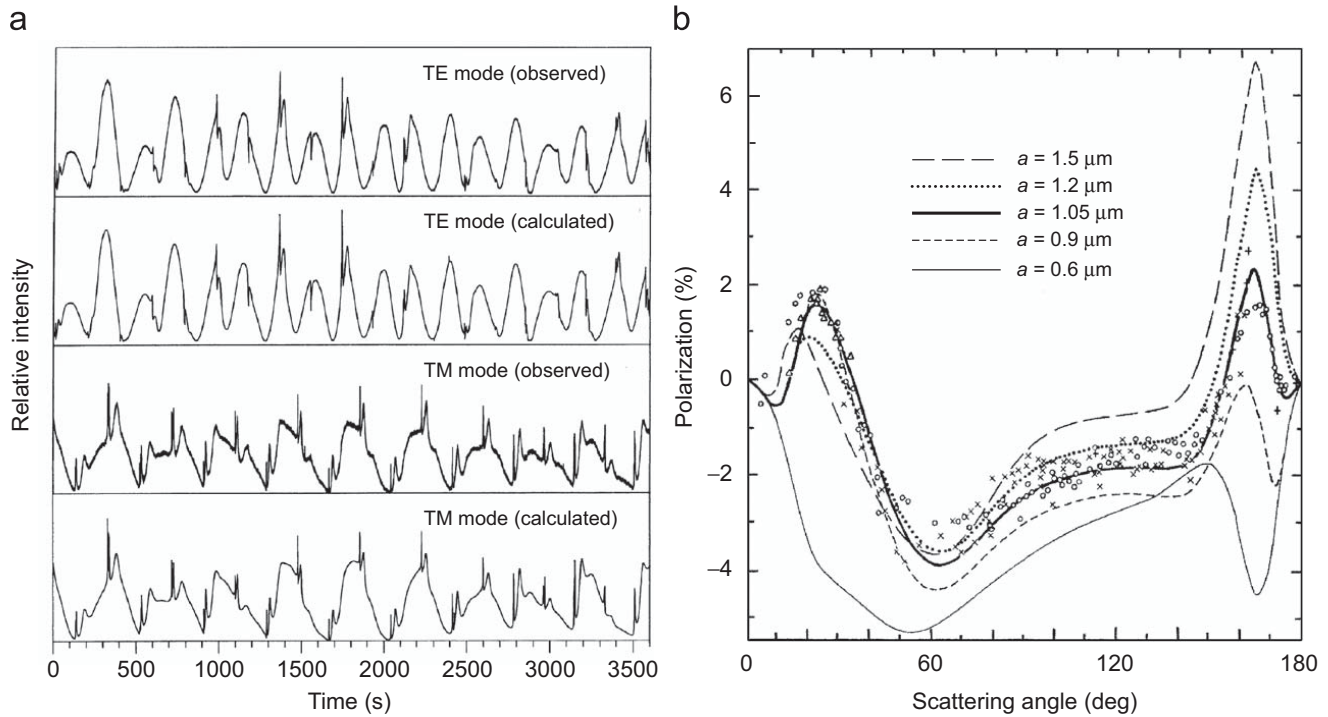
Fig. 5. (a) Comparison of observed and computed scattering-intensity spectra for a gradually evaporating glycerol droplet at scattering angles 88.54° (TE mode) and 96.44° (TM mode) (after [73]). (b) Observations of the polarization of sunlight reflected by Venus in the visual wavelength region (symbols) and theoretical computations at 550 nm wavelength (curves). The theoretical results are based on a model of cloud particles in the form of nonabsorbing spherical droplets with a relative refractive index of 1.44 and an effective variance of the droplet size distribution of 0.07. The different curves show the variability of polarization with the variation of the effective radius of the size distribution *a*.
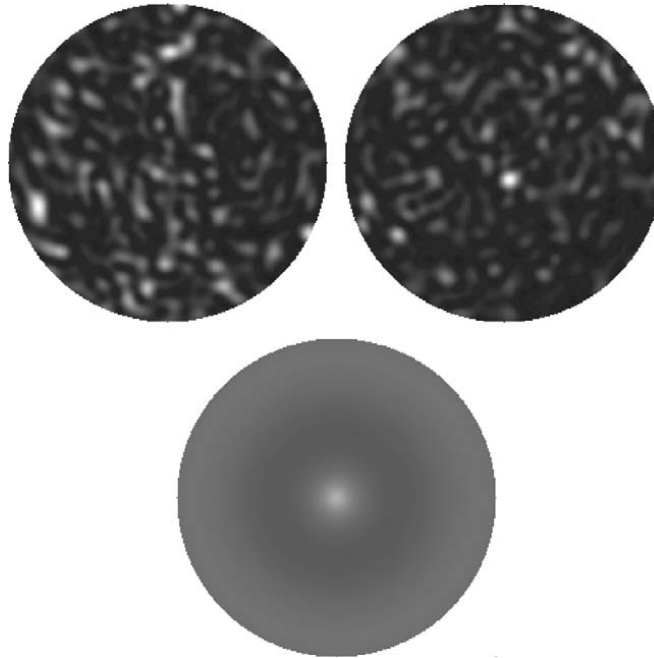

Fig. 6. Theoretical angular distribution of backscattered intensity in the far-field zone of a large spherical volume with a size parameter $k_1R = 40$ (where R is the volume radius) filled with 80 spherical particles. All particles have a fixed relative refractive index of 1.32 and a fixed radius *a* such that $k_1a = 4$ (after [64]). The upper two diagrams correspond to two different fixed particle configurations, whereas the bottom diagram is obtained by averaging over all particle positions inside the spherical volume. In all cases the volume is illuminated by a plane electromagnetic wave circularly polarized in the counterclockwise sense when viewed in the direction of propagation. The exact backscattering direction is in the center of the diagrams.
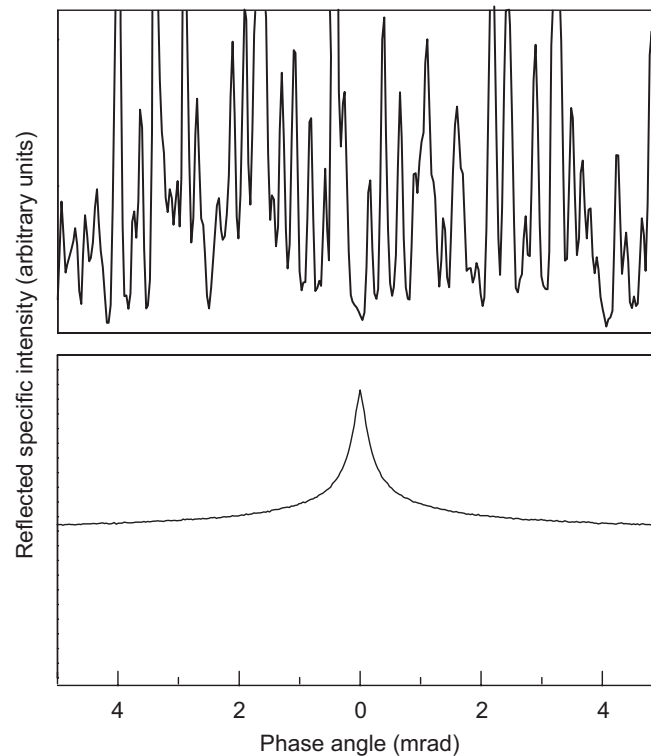
Fig. 7. The upper diagram shows a typical experimental plot of the angular distribution of backscattered specific intensity for one configuration of a particulate sample illuminated by a coherent source of light such as a laser beam. The bottom diagram depicts the angular distribution of backscattered specific intensity after configurational averaging (after [79]). The phase angle is defined as the angle between the source of light and the detector as viewed from the sample.

cloud particle chemical composition as a concentrated (76% by weight) aqueous solution of sulfuric acid. This spectacular result of planetary remote sensing coupled with the microphysical approach to radiative transfer is yet another striking demonstration of the validity and practical power of Mie's classical concept of electromagnetic scattering by particles.

An important prediction of the theory of electromagnetic scattering by a random group of sparsely distributed particles is the so-called effect of coherent backscattering (otherwise known as weak localization of electromagnetic waves; see, e.g., [62–64] and references therein). This effect cannot be singled out in the intensity of light scattered by a fixed multi-particle configuration (see the upper panels of Figs. 6 and 7) but emerges upon statistical averaging over particle positions as a narrow intensity peak centered at the exact backscattering direction (see the bottom panels of Figs. 6 and 7). The very fact that this interference effect appears in both the results of numerically exact computer solutions of the Maxwell equations (Fig. 6) and the results of controlled laboratory measurements (Fig. 7) serves as an additional validation of the classical concept of electromagnetic scattering.

6. "Photonic" confusion

The "photonic" interpretation of single and multiple light scattering illustrated in Fig. 1 has its roots in Albert Einstein's 1905 paper on the photoelectric effect. Specifically, he suggested that "the energy of a light ray spreading out from a point source is not continuously distributed over an increasing space but consists of a finite number of energy quanta which are localized at points in space, which move without dividing, and which can only be produced and absorbed as complete units" [80, p. 368]. It is these phenomenological photons that allegedly form the incident beam in Figs. 1a and b and then change the direction of their flight upon scattering on the macroscopic particle(s) or disappear inside the particle(s) due to absorption.

Although the obsolete phenomenological nature of Einstein's light quanta [81] becomes obvious upon opening virtually any advanced textbook on quantum electrodynamics (e.g., [82–87]), the lasting misinterpretation of photons as localized particles of light is kept flourishing by ignorant authors of many school and college textbooks on physics. A typical example is [88], where one can find the following false statement: "It was found that an electromagnetic wave consists of tiny localized bundles of energy. These bundles, or quanta of light, have come to be called photons" (p. 125). Another wrong statement can be found on p. 139: "Today all physicists accept that the photoelectric effect, the Compton effect, and numerous other experiments demonstrate beyond doubt the particle nature of light." Finally, on p. 140 one reads: "We can understand a large part of modern physics, armed with just the basic facts of quantum radiation theory, as summarized in

the two equations":

$$E = h\nu \quad \text{and} \quad p = \frac{h}{\lambda}, \tag{7}$$

where E, ν, p, and λ are the energy, frequency, momentum, and wavelength of the photon, respectively, and h is the Planck constant. It is obvious that armed with this notion of "photons" one would indeed take for granted the definition of electromagnetic radiation as a "shower of particles" [49, p. 32] and the photonic interpretation of electromagnetic scattering as depicted in Fig. 1.

However, the problems with the photonic interpretation of electromagnetic scattering by macroscopic particles are many. Indeed, Fig. 1a implies accepting that light propagates as a stream of photons before it reaches the particle, decides to become a wave when it impinges upon the particle and thereby generates a multitude of spectacular effects such as the diffraction pattern, rainbows, glory, MDRs, etc., and then changes its mind again upon leaving the particle and resumes its journey in the form of a stream of photons. This willful juggling with waves and photons is usually justified by a reference to the so-called "wave–particle duality of light", despite the fact that this alleged duality was discarded seven decades ago following the development of quantum electrodynamics. However, the physical insolvency of willfully thrusting a mode of behavior (i.e., a "wave" or a "particle") upon electromagnetic radiation instead of deriving it from first principles is rather obvious.

First of all it is the interaction of light with matter that most often requires quantization of energy, not the phenomenon of light propagation.

Second, actual physical photons appear as the result of quantization of the *microscopic* electromagnetic field, and so arbitrarily calling an object a "photon" does not make it physically real unless the electromagnetic field is quantized explicitly. However, the explicit quantization of the microscopic electromagnetic field in the presence of a vast number of elementary particles forming a macroscopic scattering object (such as a cloud droplet) is virtually impossible and, hence, has never been done.

Third, it takes consulting a standard textbook on quantum electrodynamics [82–85] to realize that the real physical photon is a quantum of a single normal mode of the microscopic electromagnetic field. As such each photon occupies the entire quantization volume. For example, a photon representing a plane electromagnetic wave occupies the entire three-dimensional space. Also, it is known from quantum electrodynamics that there is no position operator for a photon and that it is impossible to introduce a photon wave function in the coordinate representation (e.g., [85, Section 2.2]). Therefore, photons are not localized particles of light. If the solution of a specific problem necessitates the quantization of the electromagnetic field then the most one can say is that the resulting photons represent a discrete character of light in that specific application but not a "particle nature" of light.

Fourth, it is well established that the alleged particle behavior of light in phenomena such as the photoelectric and Compton effects can be explained quantitatively in terms of the semi-classical approach wherein the electromagnetic field is not quantized and is described by the classical microscopic Maxwell equations [89–92]. For some reasons this fact has not been widely publicized and is never mentioned in school and college textbooks in physics.

Fifth, the "photonic" interpretation of scattering shown in Fig. 1a is inconsistent with Eqs. (1) and (2). Indeed, Fig. 1a implies that the incident beam is modified by the scattering object via the removal of photons from the beam: the initial number of photons in the beam is reduced after it has passed the object. However, Eqs. (1) and (2) imply that the incident beam and its energy are not modified whatsoever by the presence of the object and are the same in the presence and in the absence of the object.

The concept of photons has been especially misused in the phenomenological treatment of radiative transfer. Indeed, a traditional phenomenological way to introduce the RTE is to describe the radiation field in terms of a "photon gas" and postulate that the latter satisfies the Boltzmann kinetic equation (see, for example, [48,93–96]). This approach is based on associating energy transport with the directional flow of localized particles of light each carrying energy of amount $h\nu$. The specific intensity of multiply scattered light is then given by $h\nu c f(\mathbf{r}, \hat{\mathbf{q}})$, where c is the speed of light and $f(\mathbf{r}, \hat{\mathbf{q}})$ is the photon distribution function such that $dS d\Omega c f(\mathbf{r}, \hat{\mathbf{q}})$ is the number of photons crossing an element of surface area dS normal to $\hat{\mathbf{q}}$ and centered at $\mathbf{r}$ in propagation directions confined to an element of solid angle $d\Omega$ centered around the unit vector $\hat{\mathbf{q}}$ per unit time.

However, quantum electrodynamics does not allow one to associate the position variable $\mathbf{r}$ with a photon and even to speak about the probability of finding a photon at a particular point in space [97]. Again, photons are not localized particles (e.g., [82, Section 4.10], [83, Section 88], and [84, Section 5.1]), which makes the expressions like "photon position", "photon path", "photon trajectory", or "local flow of photons" physically meaningless. It is, thus, impossible to define $f(\mathbf{r}, \hat{\mathbf{q}})$ as a function of photon coordinates and claim that it satisfies a Boltzmann transport equation reducible to the RTE.

Similarly, Bohren and Clothiaux [45, p. 253] claim that "the photon language is the natural one for discussing the radiative transfer theory." They "look upon photons as discrete blobs of energy without phases" and allege that their "radiative transfer theory is a theory of multiple scattering of photons rather than waves." However, they completely miss two fundamental points [63,64]:

- the RTE and the effect of coherent backscattering are inseparable and are direct consequences of averaging over time the speckle pattern generated by a multi-particle group; and
- the speckle pattern cannot be described in terms of "discrete blobs of energy without phases".

Another fundamental problem with the above "photonic" approach is that it remains absolutely unclear why the phase and extinction matrices entering the RTE and, by design, controlling the behavior of localized particles of light are still defined in the framework of classical electromagnetic scattering of waves and are computed by solving the macroscopic Maxwell equations using the Mie theory or one of its generalizations.

Yet another problem with the "photonic" interpretation of multiple scattering is that it implies that the incident "stream of photons" is exponentially attenuated as it "propagates" through a turbid medium (Fig. 1b). However, as we have already emphasized, in reality the incident plane electromagnetic wave is not modified by scattering and absorption but rather remains unchanged. What is attenuated exponentially is the time-independent so-called coherent field $\mathbf{E}_c(\mathbf{r})$ [63,98,99]. The latter is obtained by

- writing the total electric field inside the medium as $\mathbf{E}_0(\mathbf{r}, t) \exp(-i\omega t)$, where the electric field amplitude $\mathbf{E}_0(\mathbf{r}, t)$ is a "slowly varying" function of time provided that significant changes in particle positions occur over time intervals much longer than the period of time-harmonic oscillations $2\pi/\omega$;
- artificially neglecting the time-harmonic factor $\exp(-i\omega t)$;
- expressing the random amplitude $\mathbf{E}_0(\mathbf{r}, t)$ as a sum of the time-independent coherent (average) field $\mathbf{E}_c(\mathbf{r})$ and a fluctuating field $\mathbf{E}_f(\mathbf{r}, t)$ caused by random changes in particle positions; and
- calculating $\mathbf{E}_c(\mathbf{r})$ as the average of $\mathbf{E}_0(\mathbf{r}, t)$ over a time interval long enough to establish full ergodicity of the medium.

This means that the coherent field is an artificial mathematical construction rather than a real time-dependent physical field. In particular, it is not a time-harmonic plane electromagnetic wave. The only reason to consider this purely mathematical quantity in the first place is that it happens to be useful in the derivation of formulas for certain optical observables in the context of the radiative transfer theory [64].

Unfortunately, the word "photon" is invoked most commonly in circumstances in which the electromagnetic field is classical and has no quantum character whatsoever. In such cases the word "photon" serves as nothing more than a catchy synonym for "light". A typical example is the widespread use of the word "photon" in descriptions of Monte Carlo procedures for the numerical solution of the RTE for turbid media (e.g., [45,100]). In reality, however, the Monte Carlo technique involves the use of arbitrary imaginary "packets" or "units" of energy rather than the actual physical photons appearing in the context of microscopic quantum electrodynamics. Therefore, the usage of the word "photon" in the context of a numerical Monte Carlo solution of the RTE is especially misleading and should be avoided.

7. Conclusions

Although more than a hundred years have passed since the publication of Mie's seminal paper, no compelling need to modify the classical concept of electromagnetic scattering by macroscopic particles has been identified and documented. This concept has been instrumental in the development of theoretical and experimental methods to treat the interaction of the electromagnetic field with macroscopic particles and particle groups as well as in the development and application of laboratory and remote-sensing particle characterization techniques of unparalleled accuracy. The use of this concept in the detailed microphysical derivation of the RTE has clarified the physical meaning of all participating quantities, made unnecessary the multiple controversial assumptions of the phenomenological approach, and established the fundamental link between the radiative transfer theory and the effect of weak localization of electromagnetic waves in discrete random media.

Fundamentally, the classical concept of electromagnetic scattering obviates the need to use the misleading "photonic" language. Contrary to what most school and college textbooks in physics may say, Newton's light corpuscles [101], Einstein's phenomenological light quanta localized at points in space [80], and Lewis's photons as atoms of light [102] have long been history and are not the real photons appearing in quantum electrodynamics. An excellent remedy to these textbooks are the thorough discussions of the concept of a photon and its history in [90,92,103]. In particular, Kidd et al. [90] boldly and justifiably assert that elementary physics textbooks would do well to drop the corpuscular photon in favor of the semi-classical treatment as the first approximation to the modern quantum electrodynamics approach. To quote Scully and Sargent [103], the concept of the photon "has its logical foundation in the quantum theory of radiation. But the 'fuzzy-ball' picture of a photon often leads to unnecessary difficulties." Some of these unnecessary difficulties were discussed in the preceding section.

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