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TRANSFER OF ELECTROMAGNETIC ENERGY IN PARTICULATE MEDIA: FROM WAVE THEORY OF SCATTERING TO BALLISTIC TRANSPORT EQUATION

Michael I. Mishchenko
NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025
mmishchenko@giss.nasa.gov

ABSTRACT

This talk provides a general discussion of the fundamental concept of electromagnetic scattering by particles and particle groups and dispels certain widespread yet profoundly confusing misconceptions of the phenomenological theory of electromagnetic energy transport.

Key Words: Radiative transfer, electromagnetic energy, polarization, scattering

1. INTRODUCTION

Since the fundamental paper by Gustav Mie [1], electromagnetic scattering by macroscopic particles (such as cloud droplets and aerosols) has been the subject of countless papers and monographs. However, the very notions of “scattering” and “multiple scattering” have seldom been defined explicitly and have usually been assumed to be intuitively obvious. It is this underlying uncertainty that can in fact be blamed for the very appearance of the phenomenological theory of radiative transfer with its fundamental “lack of connection to mainland physics” [2].

The main objective of this talk is to clarify the concepts of single and multiple scattering of light by macroscopic particles and particle groups and trace how they shape the unified microphysical theory of radiative transfer and weak localization. Following Mie [1], we will consistently operate in the framework of macroscopic Maxwell’s electromagnetics. This approach will demonstrate how one can avoid completely the need for the profoundly confusing notion of photons as localized particles of light.

2. PHOTONS

The ballistic nature of the equation describing the transport of neutrons can be directly attributed to the fact that neutrons are quantum-mechanical particles. A similar mathematical structure of the equation describing the diffusion of electromagnetic energy in discrete random media is also frequently attributed to the so-called wave–particle duality of light allegedly causing electromagnetic energy to behave as a “stream of photons”.

It is often asserted that upon collision with an atmospheric particle, a photon can be either absorbed or scattered. Scattering is then defined as a random choice of new direction of propagation for the photon. This portrayal of scattering as a “collision” of a light corpuscle with

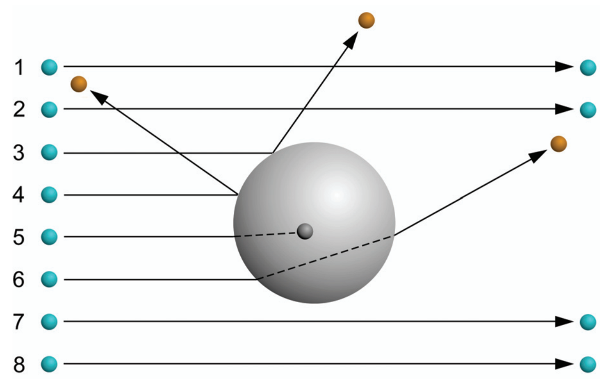

Fig. 1. Interaction of a “photonic beam” with a macroscopic particle.

a particle cloud droplet followed by the corpuscle changing the direction of flight (Fig. 1) is succinctly summarized by Bohren and Clothiaux [3]: “an incident photon becomes a scattered photon.”

In apparent agreement with this picture of electromagnetic scattering, Thomas and Stamnes [4] discuss 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. 2). The radiative transfer equation (RTE) is then introduced by describing the radiation field in

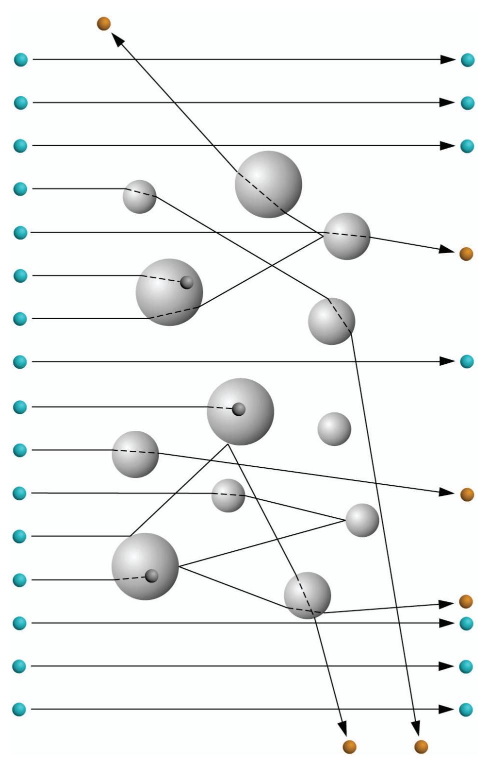

Fig. 2. Multiple scattering of a “photonic beam” by a group of particles.

terms of a "photon gas" and asserting that the latter must satisfy the Boltzmann kinetic equation. Similarly, Bohren and Clothiaux [3] 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."

The "photonic" interpretation of single and multiple light scattering illustrated in Figs. 1 and 2 has its roots in Einstein's 1905 paper on the photoelectric effect. Although the obsolete phenomenological nature of Einstein's light quanta localized in space becomes obvious upon opening virtually any advanced textbook on quantum electrodynamics, 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 [5], where one can find the following wrong 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 erroneous 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": $E = h\nu$ and $p = h/\lambda$, 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" and the photonic interpretation of electromagnetic scattering as depicted in Figs. 1 and 2.

However, the problems with the photonic interpretation of electromagnetic scattering by macroscopic particles are profound. Indeed, Fig. 1 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, morphology-dependent resonances, 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 notorious "wave–particle duality of light", despite the fact that the latter 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.

The concept of photons has been especially misused in the phenomenological treatment of radiative transfer (RT). Indeed, as we have mentioned, a popular phenomenological way to introduce the RT equation (RTE) is to describe the radiation field in terms of a "photon gas" and postulate that the latter satisfies the Boltzmann kinetic equation. 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 $\mathrm{d}S\,\mathrm{d}\Omega\, c\, f(\mathbf{r}, \hat{\mathbf{q}})$ is the number of photons crossing an element of surface area $\mathrm{d}S$ normal to $\hat{\mathbf{q}}$ and centered at $\mathbf{r}$ in propagation directions confined to an element of solid angle $\mathrm{d}\Omega$ centered around the unit vector $\hat{\mathbf{q}}$ per unit time. However, quantum electrodynamics does not allow one to associate the position variable **r** with a photon and even to speak about the probability of finding a photon at a particular point in space. Again, photons are not localized particles of light, which makes the expressions like “photon position”, “photon path”, “photon trajectory”, or “local flow of photons” physically irrelevant. As a consequence, it is 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 [3, 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 [6,7]:

- the RTE and the effect of weak localization of electromagnetic waves in discrete random media 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, e.g., 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 particulate medium (Fig. 2). In reality, however, 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 $\mathbf{E}_c(\mathbf{r})$. The latter is obtained by

- writing the total electric field inside the medium as $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r}, t)\exp(-\mathrm{i}\omega t),$ where **r** is observation point, t is time, ω is the angular frequency, 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 $\exp(-\mathrm{i}\omega t)$;
- expressing the random amplitude $\mathbf{E}_0(\mathbf{r}, t)$ as a superposition of the time-independent coherent 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 even consider the coherent field 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 RT theory [6].

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” or “electromagnetic radtaion”. 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 particulate media (e.g., [8]). 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.

3. MISCOPHYSICAL APPROACH TO RADIATIVE TRANSFER

Irrespective of how realistic various phenomenological accounts of RT may look, they inevitably fall apart upon a careful analysis of their physical foundation. Furthermore, as we have already mentioned, the phenomenological accounts completely ignore the inherent link between RT and the effect of WL. Most fundamentally, they conceal the indisputable fact that as long as scattering occurs without frequency redistribution and the particles are macroscopic and can be characterized by a refractive index, the RTE describes multiple scattering of classical electromagnetic waves and, as such, must be derived directly from the macroscopic Maxwell equations. Such a derivation has recently been published [6] and can be used to clarify the role and physical meaning of the various quantities entering the RTE, trace the link between the theories of RT and WL, cross-examine the terminologies used in the traditional phenomenological and the new microphysical approaches, and identify and correct numerous misconceptions of the phenomenological approach.

In modern physical terms, the Mie theory as well as its various generalizations belong in the realm of so-called frequency-domain macroscopic electromagnetics. This means that all fields and sources of fields are assumed to vary in time harmonically. 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. 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. However, in the presence of a particle the electromagnetic field differs from that corresponding to the unbounded homogeneous space. 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. 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: $\mathbf{E}(\mathbf{r}) = \mathbf{E}^{\text{inc}}(\mathbf{r}) + \mathbf{E}^{\text{sca}}(\mathbf{r})$, $\mathbf{H}(\mathbf{r}) = \mathbf{H}^{\text{inc}}(\mathbf{r}) + \mathbf{H}^{\text{sca}}(\mathbf{r})$, where 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. 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.

The mathematical origin of the “multiple-scattering” terminology in the RT theory can be traced back to the frequency-domain so-called Foldy–Lax equations applied to a fixed scattering object composed of N geometrically non-overlapping particles. These equations follow directly from the Maxwell equations and can be used to derive the following “order-of-scattering expansion” of the total electric field at an observation point:

$$E = E^{\text{inc}} + E^{\text{sca}}, \tag{1}$$

$$E^{\text{sca}} = \sum_{i=1}^{N} \hat{G}\hat{T}_i E^{\text{inc}} + \sum_{\substack{i=1 \\ j(\neq i)=1}}^{N} \hat{G}\hat{T}_i \hat{G}\hat{T}_j E^{\text{inc}} + \sum_{\substack{i=1 \\ j(\neq i)=1 \\ l(\neq j)=1}}^{N} \hat{G}\hat{T}_i \hat{G}\hat{T}_j \hat{G}\hat{T}_l E^{\text{inc}} + \cdots, \tag{2}$$

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 $\vec{\vec{T}}_i(\mathbf{r}', \mathbf{r}'')$ of particle i, and

$$\hat{G}\hat{T}_j E = \int_{V_j} \mathrm{d}\mathbf{r}' \vec{\vec{G}}(\mathbf{r}, \mathbf{r}') \cdot \int_{V_j} \mathrm{d}\mathbf{r}'' \vec{\vec{T}}_j(\mathbf{r}', \mathbf{r}'') \cdot \mathbf{E}(\mathbf{r}''), \tag{3}$$

where V_j is the interior volume of particle j. The dyadic transition operators are independent of each other, and each of them serves as a complete individual electromagnetic identifier of the corresponding particle. It is, therefore, quite tempting to interpret $\hat{G}\hat{T}_i E^{\text{inc}}$ as the partial scattered field at the observation point generated by particle i in response to the “excitation” by the incident field only, $\hat{G}\hat{T}_i \hat{G}\hat{T}_j E^{\text{inc}}$ as the partial field generated by the same particle but 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. (1) then represents the unscattered (i.e., incident) field.

The use of the “multiple scattering” terminology may be a convenient and compact way of illustrating the numerous consequences of the Foldy–Lax equations, in particular the microphysical theories of RT and WL. However, it directly follows from the Flody–Lax equations that all mutual particle–particle “excitations” occur simultaneously and are not temporally discrete and ordered events. Thus, the concept of multiple scattering is as much a purely mathematical abstraction as the incident and scattered fields.

The RT theory is an expressly near-field theory which follows directly from the Foldy–equations and allows one to model the response of detectors of electromagnetic energy located inside or relatively close to a random multi-particle scattering object. Among the conditions of applicability of the RTE are the asymptotic requirement $N \to \infty$, the “low-density” requirement according to which every particle must be located in the far-field zones of all the other particles, and the assumption that the scattering signal is accumulated over a time interval long enough to allow the particles move throughout the scattering volume and establish full ergodicty of the particle group. The classical integro-differential form of the RTE reads

$$\hat{\mathbf{q}} \cdot \nabla \tilde{\mathbf{I}}(\mathbf{r}, \hat{\mathbf{q}}) = -n_0 \langle \mathbf{K}_1(\hat{\mathbf{q}}) \rangle_\xi \tilde{\mathbf{I}}(\mathbf{r}, \hat{\mathbf{q}}) + n_0 \int_{4\pi} d\hat{\mathbf{q}}' \langle \mathbf{Z}_1(\hat{\mathbf{q}}, \hat{\mathbf{q}}') \rangle_\xi \tilde{\mathbf{I}}(\mathbf{r}, \hat{\mathbf{q}}'), \tag{4}$$

where n_0 is the average particle number density, $d\hat{\mathbf{q}}'$ is an elementary solid angle centered around the unit vector $\hat{\mathbf{q}}'$, $\langle \mathbf{Z}_1(\hat{\mathbf{q}}, \hat{\mathbf{q}}') \rangle_\xi$ and $\langle \mathbf{K}_1(\hat{\mathbf{q}}) \rangle_\xi$ are the single-particle phase and extinction matrix, respectively, computed with respect to the particle-centered coordinate system and averaged over all physically realizable particle states in the group, and $\tilde{\mathbf{I}}(\mathbf{r}, \hat{\mathbf{q}})$ is the so-called specific Stokes column vector having the dimension of monochromatic radiance. The latter describes the *time-averaged* (or *ensemble-averaged*) radiometric and polarimetric characteristics of electromagnetic radiation propagating in the direction of the unit vector $\hat{\mathbf{q}}$ at the observation point $\mathbf{r}$.

The RTE does not describe specifically the effect of WL and in particular its main manifestation in the form of a narrow intensity peak centered at the exact backscattering direction. Fortunately, this effect is virtually unobservable for such large rarified objects as clouds, unless the measurements are performed with a monostatic lidar/radar and the multiple-scattering contribution to the backscattered signal is significant. It is easier to observe various manifestations of WL for densely packed particulate media. The rigorous general microphysical theory of WL is extremely complex and is still hardly applicable to analyses of actual experimental data. However, in the case of low-density particulate media all manifestations of WL in the exact backscattering direction can be described quantitatively in terms of a solution of the RTE. This result ensures the applicability of the RT theory to analyses of perfectly monostatic observations.

The power of the microphysical approach to RT has recently been exploited to derive the generalized form of the RTE applicable to a large group of sparsely, randomly, and uniformly distributed particles embedded in an absorbing host medium [9,10]. Indeed, the specific form of the RTE and the physical meaning of the participating quantities in the important case of an absorbing host medium have been the subject of a lasting controversy. This controversy can largely be attributed to the conflicting outcomes of several phenomenological studies based on the common assumption that the RTE exists even when the host medium is absorbing and has the standard mathematical structure, but needs modified participating quantities.

As always, the best and most physically straightforward way to resolve such a controversy is to (i) adhere only to quantities that can be measured directly and (ii) use a microphysical rather than a phenomenological approach and derive rather than guess the final equations. In this particular case the microphysical approach should be based directly on the macroscopic Maxwell equations in much the same way as it has been done for the case of a non-absorbing host medium. Specifically, the reliance on the macroscopic Maxwell equations allows one to perform a general and systematic analysis of the problem of single electromagnetic scattering by an arbitrary finite fixed object embedded in an absorbing, homogeneous, isotropic, and unbounded medium. The corresponding generalized volume integral equation can then used to derive generalized formulas of the far-field approximation and introduce direct optical observables such as the phase and extinction matrices. Furthermore, it can also be used to derive generalized Foldy–Lax equations and their order-of-scattering form for a multi-particle group. Finally, the far-field version of the Foldy–Lax equations can be used to derive the transport equation for the coherent field generated by a large group of sparsely, randomly, and uniformly distributed particles as well as the full RTE.

Thus, the microphysical approach allows one to derive the RTE rather than to guess it in the practically important case of an absorbing host medium. This yields unambiguous and definitive analytical expressions for the participating quantities and ends the lasting controversy caused by the use of different heuristic approaches. The resulting RTE is remarkably similar to that in the case of a non-absorbing host medium, Eq. (4), and, in fact, has an intuitively obvious structure. It is straightforward to use the integral-equation counterpart of the RTE in order to demonstrate that the physical meaning of the elements of the full, diffuse, and coherent Stokes column vectors entering the RTE is exactly the same as in the case of a non-absorbing host medium. In particular, the solution of the RTE yields all quantities necessary to evaluate the electromagnetic energy budget of the entire scattering volume filled with particles (or any part of it) or to describe the time-averaged response of a well collimated polarization-sensitive detector of electromagnetic energy placed inside or outside the volume. Furthermore, the generalized RTE also remains applicable in the case of external illumination in the form of a parallel quasi-monochromatic beam of light.

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