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"Independent" and "dependent" scattering by particles in a multi-particle group

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Abstract: The terms "independent" and "dependent" scattering are ubiquitous in the phenomenological discipline of light scattering by particulate media. Yet there is a wide range of *ad hoc* definitions of these terms, many of which are vague and conceptually inconsequential. In this paper we perform a first-principles analysis of these terms based on the rigorous volume-integral-equation formulation of electromagnetic scattering. We argue that scattering by a multi-particle group can be called independent if certain optical observables for the entire group can be expressed in appropriate single-particle observables. Otherwise one deals with the dependent scattering regime. The prime (and perhaps the only) examples of independent scattering are scattering scenarios described by the first-order-scattering approximation and the first-principles radiative transfer theory.

1. Introduction

Ever since the publication of the classical treatise by Hendrik C. van de Hulst [1] in 1957, the notions of "independent" and "dependent" scattering have permeated applied publications dealing with frequency-domain electromagnetic scattering by particulate media (see, e.g., Refs. [2–37] and numerous references therein). However, the majority of definitions of these notions are quite vague and often range from being inconsequential to being outright wrong (a typical example is the invocation on page 3 of Ref. [38] of a scenario wherein "the scattering particle is unaffected by the presence of neighboring particles"). The common trait of such definitions is that they are based on qualitative *ad hoc* arguments rather than emerge as direct corollaries of macroscopic Maxwell's electromagnetics. It is therefore essential to perform a first-principles analysis of these notions starting from an explicit formulation of the frequency-domain Maxwell equations for a morphologically complex scattering object in the form of a group of N non-overlapping volumes called particles.

It is not the purpose of this tutorial paper to discuss specifically all the numerous definitions of independent and dependent scattering regimes encountered in the literature. Indeed, that would largely amount to deciphering what the authors may have wanted to say rather than analyzing what they have stated explicitly. Instead, the main objective of this paper is to advance the premise according to which scattering by a multi-particle group is *independent* if certain optical observables (i.e., appropriately defined second moments in the electromagnetic field) for the entire group can be expressed (explicitly or implicitly) in appropriate single-particle observables. Otherwise the scattering by the multi-particle group is *dependent*. The purpose of the following sections is to give a systematic and self-consistent justification of this premise and give examples of independent and dependent scattering scenarios.

In lieu of using the standard differential-equation formalism of electromagnetic scattering, we build our analysis on the mathematically equivalent volume-integral-equation (VIE) formulation. The unique advantage of the latter is that it naturally leads to the introduction of mutually independent individual-particle transition operators and yields a rigorous expression of the field scattered by an N-particle group in terms of the N single-particle transition operators. As such, this formalism allows one to bring the notion of individual-particle scattering into the consideration of electromagnetic scattering by the entire multi-particle object. Furthermore, it applies to a very wide range of particle morphologies. Although the

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majority of theoretical results used below are known, they still need to be organized in a particular way to make our reasoning and conclusions unequivocal.

2. Scattering problem

The state-of-the art of the VIE formulation of frequency-domain electromagnetic scattering by an arbitrary fixed object has recently been summarized in Refs. [39–41]. Therefore, we recapitulate here only the most essential results while using exactly the same terminology and notation. Throughout the paper, we imply (and suppress) the monochromatic $\exp(-\mathrm{i}\omega t)$ dependence of all fields, where $\mathrm{i}=(-1)^{1/2}$, ω is the angular frequency, and t is time.

Consider an *arbitrary fixed finite object* embedded in an unbounded medium that is assumed to be homogeneous, linear, isotropic, non-magnetic, and (for simplicity) nonabsorbing (Fig. 1). In general, the scattering object is an arbitrary finite group of $1 \le N < \infty$ non-overlapping components made of nonmagnetic isotropic materials, including those with edges, corners, and intersecting internal interfaces [40]. We assume that the object is subjected to an impressed incident electromagnetic field $\mathbf{E}^{\text{inc}}(\mathbf{r})$ in the form of a source-free solution of the frequency-domain Maxwell equations for an unbounded homogeneous space [41], where the position vector $\mathbf{r}$ connects the origin O of the laboratory coordinate system and the observation point.

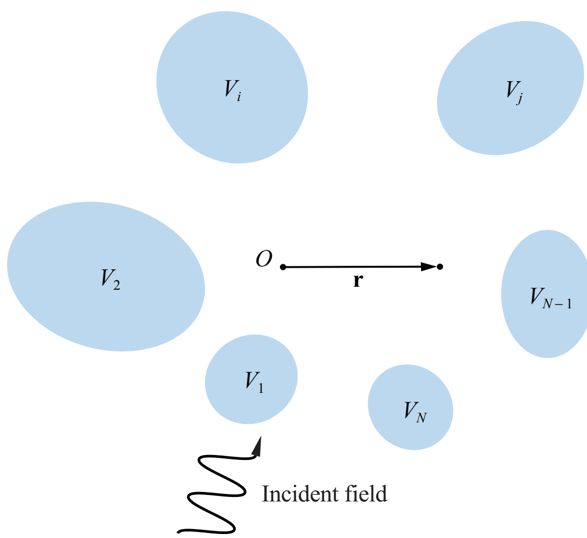

Fig. 1. Electromagnetic scattering by an arbitrary fixed finite object.

The total field everywhere in the three-dimensional space $\mathbb{R}^3$ can be represented as the sum of the incident and so-called scattered fields:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^{\text{inc}}(\mathbf{r}) + \mathbf{E}^{\text{sca}}(\mathbf{r}), \quad \mathbf{r} \in \mathbb{R}^3, \tag{1}$$

where the scattered field can be expressed in terms of the incident field and a fundamental quantity $\vec{\vec{T}}(\mathbf{r},\mathbf{r}')$, called the transition dyadic, as follows:

$$\mathbf{E}^{\text{sca}}(\mathbf{r}) = \int_{V_{\text{INT}}} \mathrm{d}^3\mathbf{r}' \,\vec{\vec{G}}(\mathbf{r},\mathbf{r}') \cdot \int_{V_{\text{INT}}} \mathrm{d}^3\mathbf{r}'' \,\vec{\vec{T}}(\mathbf{r}',\mathbf{r}'') \cdot \mathbf{E}^{\text{inc}}(\mathbf{r}''). \tag{2}$$

In the latter formula, $\vec{\vec{G}}(\mathbf{r},\mathbf{r}')$ is the free-space dyadic Green's function and V_{INT} is the cumulative volume of the object's interior, i.e., the union of the N component volumes V_{INT}^i :

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$$V_{\mathrm{INT}} \triangleq \bigcup_{i=1}^{N} V_{\mathrm{INT}}^{i}. \tag{3}$$

The free-space dyadic Green's function is a purely mathematical entity completely independent of the scattering object, whereas the transition dyadic is the solution of the following linear integral equation:

$$\ddot{T}(\mathbf{r},\mathbf{r}') = U(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}')\ddot{I} + U(\mathbf{r})\int_{V_{\mathrm{INT}}} \mathrm{d}^3\mathbf{r}''\ddot{G}(\mathbf{r},\mathbf{r}'')\cdot\ddot{T}(\mathbf{r}'',\ \mathbf{r}'),\ \ \mathbf{r},\mathbf{r}' \in V_{\mathrm{INT}}, \tag{4}$$

where

$$U(\mathbf{r}) \triangleq \begin{cases} 0, & \mathbf{r} \in \mathbb{R}^3 \setminus V_{\mathrm{INT}}, \\ \omega^2\varepsilon_2(\mathbf{r})\mu_0 - k_1^2, & \mathbf{r} \in V_{\mathrm{INT}} \end{cases} \tag{5}$$

is the potential function, $\ddot{I}$ is the identity dyadic, and $\varepsilon_2(\mathbf{r})$ is the complex permittivity of the scattering object. Furthermore, $k_1 \triangleq \omega(\varepsilon_1\mu_0)^{1/2}$ is the wave number, where ε_1 is the real-valued permittivity of the host medium and μ_0 is the magnetic permeability of a vacuum.

It is important to recognize the ultimate generality of Eq. (2) in that it is valid for any impressed incident field. Furthermore, $\ddot{T}(\mathbf{r},\mathbf{r}')$ is fundamentally independent of $\mathbf{E}^{\mathrm{inc}}(\mathbf{r})$ and is fully defined by the spatial distribution of the object's complex permittivity throughout the interior volume V_{INT} with respect to the laboratory coordinate system. As such, the transition dyadic can be considered a unique and universal "scattering ID" of the object.

To facilitate the following discussion, let us (i) extrapolate the definition of the transition dyadic to the entire space by assuming that $\ddot{T}(\mathbf{r},\mathbf{r}') = \ddot{0}$ unless $\mathbf{r} \in V_{\mathrm{INT}}$ and $\mathbf{r}' \in V_{\mathrm{INT}}$, where $\ddot{0}$ is a zero dyadic; (ii) define the potential dyadic according to $\ddot{U}(\mathbf{r},\mathbf{r}') \triangleq U(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}')\ddot{I}$, where $\delta(\mathbf{r})$ is the three-dimensional delta function; and (iii) introduce compact integral-operator notation according to

$$\hat{B}E \triangleq \int_{\mathbb{R}^3} \mathrm{d}^3\mathbf{r}'\,\ddot{B}(\mathbf{r},\mathbf{r}')\cdot\mathbf{E}(\mathbf{r}'),\quad \left(\hat{B}\hat{C}\right)E \triangleq \hat{B}\left(\hat{C}E\right), \tag{6}$$

(see Ref. [41]. for details). Then Eqs. (1), (2), and (4) can be re-written as follows:

$$E = E^{\mathrm{inc}} + E^{\mathrm{sca}} = E^{\mathrm{inc}} + \hat{G}\hat{T}E^{\mathrm{inc}}, \tag{7}$$

$$\hat{T} = \hat{U} + \hat{U}\hat{G}\hat{T}, \tag{8}$$

where $\hat{T}$ can be referred to as the transition operator of the scattering object.

3. Order-of-scattering expansion

A fundamental feature of the differential macroscopic Maxwell equations as well as of the mathematically equivalent VIE formulation summarized in the preceding section is that an object immersed in a homogeneous unbounded medium is treated by definition as a single, unified scatterer irrespective of its actual morphology. Hence the total-volume integration domains in Eqs. (2) and (4). However, the very feasibility of discussing independent and dependent scattering regimes rests on the assumption that it is somehow possible to incorporate the notion of individual-particle scattering if the object is a multi-particle group. And indeed, the decisive advantage of the VIE formulation over the differential-equation formalism is that it allows one to make explicit use of the fact that the entire scattering object can be represented geometrically as a union of N distinct non-overlapping partial volumes

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according to Fig. 1 and Eq. (3). In what follows, we will refer to these partial volumes as "particles" and to the entire scattering object as an "*N*-particle group."

The first step is to notice that the potential function of the entire object given by Eq. (5) can be rewritten as the sum of the individual-particle potential functions $\ddot{U}_i(\mathbf{r},\mathbf{r}')$:

$$\ddot{U}(\mathbf{r},\mathbf{r}') = \sum_{i=1}^{N} \ddot{U}_i(\mathbf{r},\mathbf{r}'), \quad \mathbf{r} \in \mathbb{R}^3, \tag{9}$$

where

$$\ddot{U}_i(\mathbf{r},\mathbf{r}') \triangleq \begin{cases} \ddot{0}, & \mathbf{r} \in \mathbb{R}^3 \setminus V_{\text{INT}}^i, \\ U(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}')\ddot{I}, & \mathbf{r} \in V_{\text{INT}}^i. \end{cases} \tag{10}$$

By definition, the individual-particle potential functions are independent of each other. The second step is to introduce the individual-particle transition operators with respect to the common laboratory reference frame as solutions of the following *N* separate integral equations:

$$\hat{T}_i = \hat{U}_i + \hat{U}_i \hat{G} \hat{T}_i, \; i = 1,...,N \tag{11}$$

[cf. Eq. (8)]. In other words, the transition operator of particle *i* (i.e., its "individual scattering ID") is computed as if all the other particles did not exist.

A key corollary of the VIE formalism [39,41] then states that the transition operator of the entire *N*-particle group is expressed in terms of the individual-particle transition operators:

$$\hat{T} = \sum_{i=1}^{N} \hat{T}_i + \sum_{\substack{i=1 \\ j(\neq i)=1}}^{N} \hat{T}_i \hat{G} \hat{T}_j + \sum_{\substack{i=1 \\ j(\neq i)=1 \\ l(\neq j)=1}}^{N} \hat{T}_i \hat{G} \hat{T}_j \hat{G} \hat{T}_l + \cdots. \tag{12}$$

Then, according to Eq. (7), the total field is given by the following "order-of-scattering" series:

$$E = E^{\text{inc}} + \sum_{i=1}^{N} \hat{G}\hat{T}_i E^{\text{inc}} + \sum_{\substack{i=1 \\ j(\neq i)=1}}^{N} \hat{G}\hat{T}_i \hat{G}\hat{T}_j E^{\text{inc}} + \sum_{\substack{i=1 \\ j(\neq i)=1 \\ l(\neq j)=1}}^{N} \hat{G}\hat{T}_i \hat{G}\hat{T}_j \hat{G}\hat{T}_l E^{\text{inc}} + \cdots. \tag{13}$$

It is Eq. (13) that embodies (even if only mathematically [39,42]) the sought concept of "splitting" the scattering by the entire *N*-particle group into "sequences of single-scattering events". Indeed, the term $\hat{G}\hat{T}_i E^{\text{inc}}$ can be interpreted as the contribution of the single scattering of the incident field by particle i [cf. Eq. (7)]; the term $\hat{G}\hat{T}_i \hat{G}\hat{T}_j E^{\text{inc}}$ is the contribution of the single scattering by particle i of the singly-scattered incident field by particle j; etc.

Given its fundamental importance, it is instructive to compare this result with the following two quotes. The first one is from page 11 of Ref. [7]: "Independent scattering can be defined as a condition whereby the scattering from a single particle in a cloud is not affected by the proximity of its neighbors." The second one is from page 388 of Ref. [28]: "If scattering by one particle is not affected by the presence of surrounding particles, we speak of independent scattering, otherwise we have dependent scattering." It is straightforward to see that should we apply these definitions to Eq. (13), we would have to conclude that electromagnetic scattering by any multi-particle group is always independent. Indeed, the single scattering by particle i is fully defined by the *i*th transition operator $\hat{T}_i$ which,

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according to Eq. (11), is completely independent of the presence of all the other particles. Presumably, this outcome is not what the definitions of independent scattering in Refs. [7,28] were meant to accomplish.

Note that the infinite series (13) does not necessarily converge in all cases. In fact, specific numerical examples of divergence have been reported in the literature [43,44]. In such cases any discussion of independent versus dependent scattering regimes becomes pointless.

4. Energy-budget and optical-characterization problems

Equations (11) and (13) show how to express the cumulative field scattered by a multi-particle group in terms of the single-particle transition operators. However, the electromagnetic field itself typically is not a directly observable quantity, at least in the optical range of frequencies. It is therefore important to recognize that in the usual context of electromagnetic scattering by particles, the only two problems of actual practical significance are as follows [39,45]:

- How to evaluate theoretically the time-averaged radiation-energy budget of a macroscopic volume of particulate medium?
- How to model theoretically the particular measurement afforded by an actual detector of electromagnetic energy flow and thereby clarify its ability to serve as (i) an energy-budget meter and/or (ii) an integral part of a diagnostic technique intended for optical characterization of a particulate medium in a laboratory, *in situ*, or remote-sensing setting?

Both problems can only be solved in terms of optical observables, i.e., specific second moments in the electromagnetic field.

It is clear that if the total field is expressed according to Eq. (13) then any second moment in the field can ultimately be expressed in terms of the individual-particle transition operators. Yet the complexity of the resulting formulas is overwhelming and hardly makes this approach more practical than the use of the primordial expression (2) based on the whole-object transition operator. Thus we should look for a way to (i) drastically simplify the nested-integral expansion (13) by converting it into a purely algebraic one, and (ii) express optical observables of the *N*-particle group directly in terms of individual-particle optical observables.

Thus, the next stage in the first-principles analysis of independent and dependent scattering regimes is to import into the picture appropriate single-particle optical observables. The first step to do that is to replace individual-particle transition operators in Eq. (13) by individual-particle far-field scattering dyadics defined in the following section.

5. Far-field scattering dyadic

Let us consider the simplest scenario wherein the entire scattering object is a single particle centered at the origin of the laboratory coordinate system, as illustrated schematically in Fig. 2. The particle is subjected to the impressed incident field in the form of a homogeneous plane electromagnetic wave propagating in the direction of the unit vector $\hat{\mathbf{n}}^{\mathrm{inc}}$:

$$\mathbf{E}^{\mathrm{inc}}(\mathbf{r}) = \exp(\mathrm{i}k_1 \hat{\mathbf{n}}^{\mathrm{inc}} \cdot \mathbf{r})\mathbf{E}_0^{\mathrm{inc}}, \quad \mathbf{E}_0^{\mathrm{inc}} \cdot \hat{\mathbf{n}}^{\mathrm{inc}} = 0. \tag{14}$$

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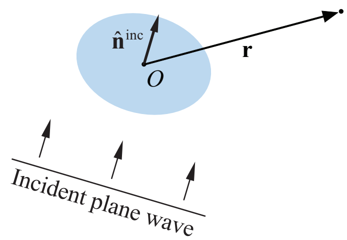

Fig. 2. Far-field scattering by a single particle.

Note that the use of a caret above an upright bold-face character to denote a unit vector should not be confused with the previous use of a caret above an italic character to denote an integral operator.

A well-known fundamental result of the theory of electromagnetic scattering by small particles [39,41,46,47] is that at a sufficiently large distance from the particle, the scattered field evolves into a transverse outgoing spherical wave:

$$\mathbf{E}^{\rm sca}(\mathbf{r}) \underset{r\to\infty}{=} g(r)\vec{A}(\hat{\mathbf{r}},\hat{\mathbf{n}}^{\rm inc})\cdot\mathbf{E}_0^{\rm inc}, \quad \mathbf{E}^{\rm sca}(\hat{\mathbf{r}})\cdot\hat{\mathbf{r}} = 0, \tag{15}$$

where $r = |\mathbf{r}|$; $\hat{\mathbf{r}} = \mathbf{r}/r$; $g(r) = \exp(\mathrm{i}k_1 r)/r$; $\vec{A}(\hat{\mathbf{r}},\hat{\mathbf{n}}^{\rm inc})$ is the so-called far-field scattering dyadic; and the radial unit vector $\hat{\mathbf{r}}$ plays the role of the scattering direction. The scattering dyadic is expressed in terms of the transition dyadic of the particle according to

$$\begin{aligned}\vec{A}(\hat{\mathbf{r}},\hat{\mathbf{n}}^{\rm inc}) &= \frac{1}{4\pi}\left(\vec{I} - \hat{\mathbf{r}}\otimes\hat{\mathbf{r}}\right)\cdot\int_{V_{\rm INT}} \mathrm{d}^3\mathbf{r}'\exp(-\mathrm{i}k_1\hat{\mathbf{r}}\cdot\mathbf{r}') \\ &\times\int_{V_{\rm INT}} \mathrm{d}^3\mathbf{r}''\vec{T}(\mathbf{r}',\mathbf{r}'')\cdot\left(\vec{I} - \hat{\mathbf{n}}^{\rm inc}\otimes\hat{\mathbf{n}}^{\rm inc}\right)\exp(\mathrm{i}k_1\hat{\mathbf{n}}^{\rm inc}\cdot\mathbf{r}''),\end{aligned} \tag{16}$$

where $V_{\rm INT}$ is the volume occupied by the particle and $\otimes$ is the dyadic product sign.

6. Simplified order-of-scattering expansion

Comparison of Eqs. (7), (13), and (15) suggests that in order to incorporate the notion of the individual-particle far-field scattering dyadic in the computation of the field scattered by the entire N-particle group, one has to assume that (i) the group is subjected to an impressed incident field in the form of a plane wave propagating in the direction $\hat{\mathbf{n}}^{\rm inc}$; (ii) each particle resides in the far zones of all the other particles; and (iii) the observation point resides in the far zone of each of the N particles (but, generally, not in the far zone of the entire group).

Indeed, let us consider, for example, the term $\hat{G}\hat{T}_i\hat{G}\hat{T}_j E^{\rm inc}$ in Eq. (13), in which $\hat{G}\hat{T}_j E^{\rm inc}$ is the result of scattering of the incident field by particle j. At the distant origin O_i of particle i, $\hat{G}\hat{T}_j E^{\rm inc}$ becomes an outgoing spherical wave centered at the origin O_j of particle j which can be considered locally plain at O_i owing to the smallness of the solid angle subtended by particle i as viewed from O_j. The subsequent application of the operator $\hat{G}\hat{T}_i$ to this quasi-plain wave then yields a field at the distant observation point which becomes an O_i-centered outgoing spherical wave. Other terms in Eq. (13) can be interpreted analogously.

To simplify Eq. (13) according to this interpretation, let us introduce the notation depicted in Fig. 3, use a caret above a vector to denote a unit vector pointing in the same direction, and denote by $\vec{A}_j$ the far-field scattering dyadic of particle j centered at O_j. We then have [39,47]:

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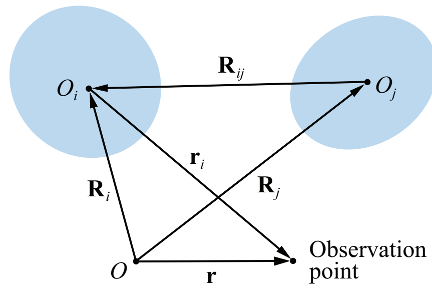

Fig. 3. Notation used in Eq. (17).

$$\begin{aligned}\mathbf{E}(\mathbf{r}) &= \mathbf{E}^{\mathrm{inc}}(\mathbf{r}) + \sum_{i=1}^{N} g(r_i)\vec{\vec{A}}_i(\hat{\mathbf{r}}_i,\hat{\mathbf{n}}^{\mathrm{inc}})\cdot\mathbf{E}^{\mathrm{inc}}(\mathbf{R}_i)\\ &+\sum_{i=1}^{N}\sum_{j(\neq i)=1}^{N} g(r_i)\vec{\vec{A}}_i(\hat{\mathbf{r}}_i,\hat{\mathbf{R}}_{ij})\cdot g(R_{ij})\vec{\vec{A}}_j(\hat{\mathbf{R}}_{ij},\hat{\mathbf{n}}^{\mathrm{inc}})\cdot\mathbf{E}^{\mathrm{inc}}(\mathbf{R}_j)\\ &+\sum_{i=1}^{N}\sum_{j(\neq i)=1}^{N}\sum_{l(\neq j)=1}^{N} g(r_i)\vec{\vec{A}}_i(\hat{\mathbf{r}}_i,\hat{\mathbf{R}}_{ij})\cdot g(R_{ij})\vec{\vec{A}}_j(\hat{\mathbf{R}}_{ij},\hat{\mathbf{R}}_{jl})\\ &\cdot g(R_{jl})\vec{\vec{A}}_l(\hat{\mathbf{R}}_{jl},\hat{\mathbf{n}}^{\mathrm{inc}})\cdot\mathbf{E}^{\mathrm{inc}}(\mathbf{R}_l)+\cdots.\end{aligned}\tag{17}$$

Importantly, the total scattered field is now a superposition of spherical waves, each one originating at the last particle of a sequence. We will see later that this enables meaningful optical measurements with well-collimated radiometers.

It is seen that all the burdensome nested integrations over particles' volumes in Eq. (13) are now gone, and the reduced order-of-scattering expansion (17) is purely algebraic. Still, the practical use of Eq. (17) remains problematic. First of all, the evaluation of optical observables for exceedingly large N (e.g., for objects such as clouds or colloidal suspensions) is hardly possible even with the use of modern computers. Second of all, the observed scattering pattern is unavoidably loaded by countless speckles that can hardly be measured let alone interpreted. In addition, optical observables for the multi-particle group still cannot be expressed in terms of individual-particle observables.

Indeed, let us assume for simplicity that N is sufficiently small that the expansion (17) can be truncated by keeping only the cumulative first-order-scattering term:

$$\mathbf{E}^{\mathrm{sca}}(\mathbf{r}) = \sum_{i=1}^{N} g(r_i)\vec{\vec{A}}_i(\hat{\mathbf{r}}_i,\hat{\mathbf{n}}^{\mathrm{inc}})\cdot\mathbf{E}^{\mathrm{inc}}(\mathbf{R}_i).\tag{18}$$

Although the observation point $\mathbf{r}$ is required to reside in the far zone of each particle in the group, we will assume that it resides in the near zone of the entire group (Fig. 4).

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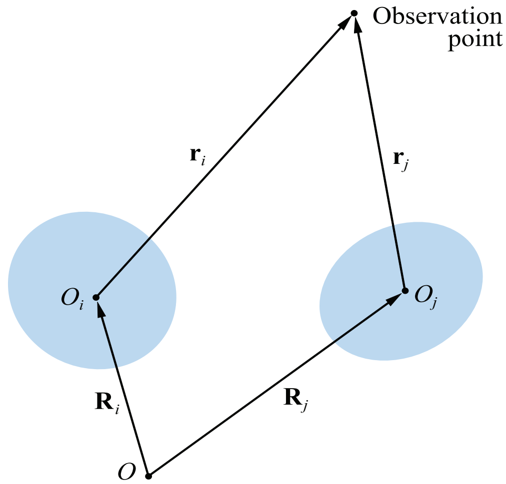

Fig. 4. Origin of the speckle pattern.

Let us consider a rather generic second moment in the scattered field called the scattering coherency dyadic and defined according to $\vec{\rho}^{\text{sca}}(\mathbf{r}) = \mathbf{E}^{\text{sca}}(\mathbf{r}) \otimes [\mathbf{E}^{\text{sca}}(\mathbf{r})]^*$, where the asterisk denotes a complex-conjugate value. Then Eq. (18) implies that

$$\vec{\rho}^{\text{sca}}(\mathbf{r}) = \sum_{i=1}^{N} \vec{\rho}_i^{\text{sca}}(\mathbf{r}) + \sum_{i=1}^{N} \sum_{j(\neq i)=1}^{N} \vec{\rho}_{ij}^{\text{sca}}(\mathbf{r}), \tag{19}$$

where

$$\vec{\rho}_i^{\text{sca}}(\mathbf{r}) = \frac{1}{r_i^2} \vec{A}_i(\hat{\mathbf{r}}_i, \hat{\mathbf{n}}^{\text{inc}}) \cdot \vec{\rho}^{\text{inc}} \cdot \left[\vec{A}_i(\hat{\mathbf{r}}_i, \hat{\mathbf{n}}^{\text{inc}}) \right]^{\text{T}*}, \tag{20}$$

are individual-particle contributions;

$$\vec{\rho}_{ij}^{\text{sca}}(\mathbf{r}) = \frac{\exp(\mathrm{i}\Delta_{ij})}{r_i r_j} \vec{A}_i(\hat{\mathbf{r}}_i, \hat{\mathbf{n}}^{\text{inc}}) \cdot \vec{\rho}^{\text{inc}} \cdot \left[\vec{A}_j(\hat{\mathbf{r}}_j, \hat{\mathbf{n}}^{\text{inc}}) \right]^{\text{T}*}, \tag{21}$$

with $\Delta_{ij} = k_1(r_i - r_j + \hat{\mathbf{n}}^{\text{inc}} \cdot \mathbf{R}_i - \hat{\mathbf{n}}^{\text{inc}} \cdot \mathbf{R}_j)$ are particle–particle pair contributions; and $\vec{\rho}^{\text{inc}} = \mathbf{E}_0^{\text{inc}} \otimes [\mathbf{E}_0^{\text{inc}}]^*$ is the coherency dyadic of the incident field. Note that in deriving Eqs. (20) and (21) we have used the dyadic identity $\left(\vec{A} \cdot \mathbf{a}\right) \otimes \left(\vec{B} \cdot \mathbf{b}\right) = \vec{A} \cdot (\mathbf{a} \otimes \mathbf{b}) \cdot \vec{B}^{\text{T}}$, where “T” stands for “transposed.”

If all the $\vec{\rho}_{ij}^{\text{sca}}(\mathbf{r})$ were equal to zero then the optical observable $\vec{\rho}^{\text{sca}}(\mathbf{r})$ of the entire group would be the sum of the mutually independent individual-particle optical observables. Indeed, the $\vec{\rho}_i^{\text{sca}}(\mathbf{r})$ for each i is computed as if all the other $N-1$ particles did not exist. In that case we could talk of the independent scattering regime.

It is obvious however that the particle–particle pair contributions $\vec{\rho}_{ij}^{\text{sca}}(\mathbf{r})$ are of the same order of magnitude as the individual-particle contributions. Furthermore, there are $N(N-1)$ particle–particle pair contributions versus only N individual-particle contributions. Therefore, there is no *a priori* reason to neglect the second (double) sum on the right-hand

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side of Eq. (19) in comparison with the first sum. In addition, the complex-exponential factor in Eq. (21) makes each $\vec{\rho}_{ij}^{\rm sca}(\mathbf{r})$ a rapidly oscillating function of the coordinates of the observation point and gives rise to a pronounced speckle pattern. This fundamental optical phenomenon is well known and is characteristic of fixed scattering objects subjected to coherent monochromatic illumination such as by a continuous-laser beam [48–50].

Thus we must conclude that in general, monochromatic electromagnetic scattering by a fixed multi-particle group is *dependent* for any $N \geq 2$ and any interparticle distances.

7. Temporal averaging, ergodicity, and ensemble averaging

Any actual measurement takes a finite amount of time. Therefore, the most obvious way to zero out the second sum on the right-hand side of Eq. (19) is to assume that all particles move incessantly during the measurement, thereby making the complex exponential in Eq. (21) a random function of time such that its temporal average vanishes: $\langle \exp(\mathrm{i}\Delta_{ij}) \rangle_t \to 0.$ Obviously, this would also serve to extinguish the speckle pattern rarely observed in practice.

The direct computation of a temporal average is highly problematic since it requires the explicit invocation of a dynamical model of the multi-particle group. Therefore, it is typical in practice to assume that the multi-particle group is ergodic and replace averaging over time by averaging over a representative ensemble of random realizations of the group in terms of varying particles' coordinates (see, e.g., Section 10.4 of Ref. [39] and Section 1.5 of Ref. [47]).

Thus, assuming that random movements of the particles imply $\langle \vec{\rho}_{ij}^{\rm sca}(\mathbf{r}) \rangle_t = \langle \vec{\rho}_{ij}^{\rm sca}(\mathbf{r}) \rangle_{\mathbf{R}} = \vec{0}$ in Eq. (19), we have

$$\langle \vec{\rho}^{\rm sca}(\mathbf{r}) \rangle_t = \langle \vec{\rho}^{\rm sca}(\mathbf{r}) \rangle_{\mathbf{R}} = \sum_{i=1}^{N} \langle \vec{\rho}_i^{\rm sca}(\mathbf{r}) \rangle_{\mathbf{R}}. \tag{22}$$

This result demonstrates that averaging over an ensemble of random configurations of a multi-particle group is a key ingredient of the independent scattering regime.

8. Single-particle extinction and phase matrices

To give examples of independent scattering regimes often encountered in practice, we need to introduce two specific types of single-particle optical observables, as follows. It is convenient to characterize a propagation direction $\hat{\mathbf{r}}$ by its polar, θ, and azimuthal, φ, angles with respect to the laboratory spherical coordinate system. This helps introduce the 2×2 amplitude scattering matrix $\mathsf{S}(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc})$ expressing the θ- and φ-components of the scattered spherical wave in terms of the θ- and φ-components of the incident plane wave:

$$\mathsf{E}^{\rm sca}(r\hat{\mathbf{r}}) \underset{r\to\infty}{=} g(r)\mathsf{S}(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc})\mathsf{E}_0^{\rm inc}, \tag{23}$$

where E denotes a two-component column formed by the θ- and φ-components of the electric field vector. It then follows from Eq. (15) that

$$\mathsf{S}(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) = \begin{bmatrix} \hat{\boldsymbol{\theta}} \cdot \vec{A}(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) \cdot \hat{\boldsymbol{\theta}}^{\rm inc} & \hat{\boldsymbol{\theta}} \cdot \vec{A}(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) \cdot \hat{\boldsymbol{\varphi}}^{\rm inc} \\ \hat{\boldsymbol{\varphi}} \cdot \vec{A}(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) \cdot \hat{\boldsymbol{\theta}}^{\rm inc} & \hat{\boldsymbol{\varphi}} \cdot \vec{A}(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) \cdot \hat{\boldsymbol{\varphi}}^{\rm inc} \end{bmatrix}. \tag{24}$$

The basic far-field individual-particle optical observables are the particle-centered real-valued 4×4 phase, $\mathsf{Z}(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc})$, and extinction, $\mathsf{K}(\hat{\mathbf{n}}^{\rm inc})$, matrices. Both are defined in terms of the elements of the particle-centered amplitude scattering matrix (24) [39,46,47]:

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$$\mathbf{K}(\hat{\mathbf{n}}^{\text{inc}}) = \frac{2\pi}{k_1}\begin{bmatrix} \text{Im}(S_{11}+S_{22}) & \text{Im}(S_{11}-S_{22}) & -\text{Im}(S_{12}+S_{21}) & \text{Re}(S_{21}-S_{12}) \\ \text{Im}(S_{11}-S_{22}) & \text{Im}(S_{11}+S_{22}) & \text{Im}(S_{21}-S_{12}) & -\text{Re}(S_{12}+S_{21}) \\ -\text{Im}(S_{12}+S_{21}) & -\text{Im}(S_{21}-S_{12}) & \text{Im}(S_{11}+S_{22}) & \text{Re}(S_{22}-S_{11}) \\ \text{Re}(S_{21}-S_{12}) & \text{Re}(S_{12}+S_{21}) & -\text{Re}(S_{22}-S_{11}) & \text{Im}(S_{11}+S_{22}) \end{bmatrix}, \tag{25}$$

$$\mathbf{Z} = \begin{bmatrix} \frac{1}{2}(|S_{11}|^2+|S_{12}|^2+|S_{21}|^2+|S_{22}|^2) & \frac{1}{2}(|S_{11}|^2-|S_{12}|^2+|S_{21}|^2-|S_{22}|^2) & -\text{Re}(S_{11}S_{12}^*+S_{22}S_{21}^*) & -\text{Im}(S_{11}S_{12}^*-S_{22}S_{21}^*) \\ \frac{1}{2}(|S_{11}|^2+|S_{12}|^2-|S_{21}|^2-|S_{22}|^2) & \frac{1}{2}(|S_{11}|^2-|S_{12}|^2-|S_{21}|^2+|S_{22}|^2) & -\text{Re}(S_{11}S_{12}^*-S_{22}S_{21}^*) & -\text{Im}(S_{11}S_{12}^*+S_{22}S_{21}^*) \\ -\text{Re}(S_{11}S_{21}^*+S_{22}S_{12}^*) & -\text{Re}(S_{11}S_{21}^*-S_{22}S_{12}^*) & \text{Re}(S_{11}S_{22}^*+S_{12}S_{21}^*) & \text{Im}(S_{11}S_{22}^*+S_{21}S_{12}^*) \\ -\text{Im}(S_{21}S_{11}^*+S_{22}S_{12}^*) & -\text{Im}(S_{21}S_{11}^*-S_{22}S_{12}^*) & \text{Im}(S_{22}S_{11}^*-S_{12}S_{21}^*) & \text{Re}(S_{22}S_{11}^*-S_{12}S_{21}^*) \end{bmatrix}. \tag{26}$$

Note that the arguments $(\hat{\mathbf{n}}^{\text{inc}}, \hat{\mathbf{n}}^{\text{inc}})$ on the right-hand side of Eq. (25) and $(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\text{inc}})$ on both sides of Eq. (26) are omitted for brevity.

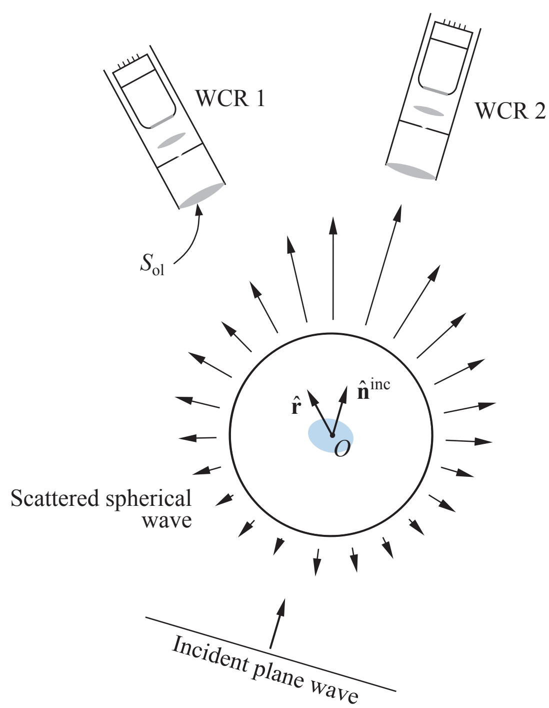

Fig. 5. The net polarized signal recorded by a WCR depends on the line of sight.

An essential practical function of the phase and extinction matrices is to quantify the reading of a specific optical instrument called the well-collimated radiometer (WCR; see Section 11.4 of Ref. [39] and Ref. [51]). The corresponding far-field measurement configuration is depicted in Fig. 5. WCR 2 has its optical axis parallel to the incidence direction and centered at the origin, while the optical axis of WCR 1 is centered at the origin in the direction of the unit vector $\hat{\mathbf{r}} \neq \hat{\mathbf{n}}^{\text{inc}}$. Let us assume that both WCRs are polarization

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sensitive and that the real-valued Stokes parameters of a transverse electromagnetic wave are defined as in Refs [39,45–47]. Then the polarized reading of WCR 1 per unit time is given by

$$\textbf{Signal1} = S_{\mathrm{ol}}\mathbf{I}^{\mathrm{sca}}(\hat{\mathbf{r}}) = \frac{S_{\mathrm{ol}}}{r^2}\mathbf{Z}(\hat{\mathbf{r}},\hat{\mathbf{n}}^{\mathrm{inc}})\mathbf{I}^{\mathrm{inc}}, \tag{27}$$

where $\mathbf{I}^{\mathrm{inc}} = [I^{\mathrm{inc}}, Q^{\mathrm{inc}}, U^{\mathrm{inc}}, V^{\mathrm{inc}}]^{\mathrm{T}}$ is the four-element Stokes column vector of the incident plane wave, $\mathbf{I}^{\mathrm{sca}}(\hat{\mathbf{r}})$ is that of the scattered spherical wave, S_{ol} is the area of the objective lens of the WCR, and r is the distance from the origin to the WCR. The corresponding polarized reading of WCR 2 per unit time is given by

$$\textbf{Signal2} = S_{\mathrm{ol}}\mathbf{I}^{\mathrm{inc}} - \mathbf{K}(\hat{\mathbf{n}}^{\mathrm{inc}})\mathbf{I}^{\mathrm{inc}} + \frac{S_{\mathrm{ol}}}{r^2}\mathbf{Z}(\hat{\mathbf{n}}^{\mathrm{inc}},\hat{\mathbf{n}}^{\mathrm{inc}})\mathbf{I}^{\mathrm{inc}}. \tag{28}$$

The extinction and phase matrices can also be used to define derivative observables called the extinction, scattering, and absorption cross sections [39,46,47]. These quantities define the electromagnetic energy budget of any finite volume of space encompassing the particle.

9. First-order-scattering approximation for a small random group of particles

An important example of the independent scattering regime is the so-called first-order-scattering approximation (FOSA) for a small sparse random group of particles (see Chapter 14 of Ref. [39]). This approximation is based on the following assumptions often encountered in laboratory and *in situ* measurements [52,53]:

- the number of particles N in the group is sufficiently small and the average distance between the particles is sufficiently large that Eq. (18) holds;
- the group is observed from a distance r much greater than any linear dimension of the volume V populated (in the statistical sense) by the group (Fig. 6);

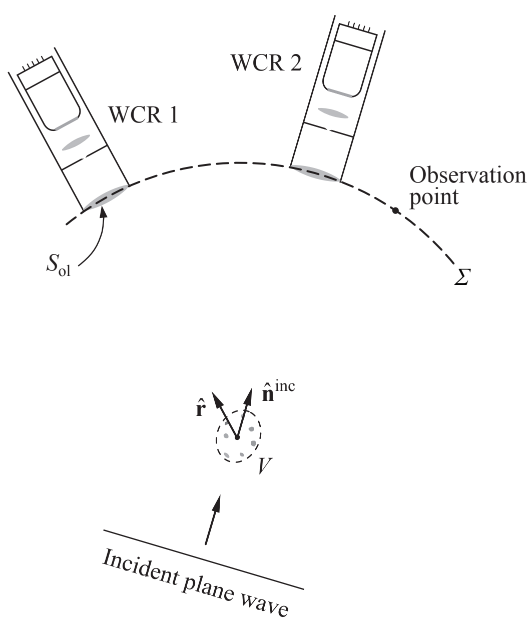

Fig. 6. A random group of N particles populating the volume V is observed from a large distance.

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- the observation point is in the near zone of the volume V yet is remote enough to be in the far zone of any particle in the group;
- the microphysical state of each particle (i.e., the combination of its shape, orientation, and size) is independent of its position and of the states and positions of all the other particles;
- the N particles are moving randomly and independently of each other throughout V; and
- the random N-particle group is fully ergodic.

Let us first consider the measurement configuration shown in Fig. 6 and paralleling that in Fig. 5. Then it can be shown [39] that the time-averaged readings of WCR 1 and WCR 2 are given by

$$\langle \textbf{Signal1} \rangle_t = \langle \textbf{Signal1} \rangle_{\mathbf{R},\xi} = \frac{S_{\rm ol}}{r^2} \sum_{i=1}^{N} \langle \mathbf{Z}^i(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) \rangle_\xi \mathbf{I}^{\rm inc}, \tag{29}$$

$$\langle \textbf{Signal2} \rangle_t = \langle \textbf{Signal2} \rangle_{\mathbf{R},\xi} = S_{\rm ol} \mathbf{I}^{\rm inc} - \sum_{i=1}^{N} \langle \mathbf{K}^i(\hat{\mathbf{n}}^{\rm inc}) \rangle_\xi \mathbf{I}^{\rm inc} + \frac{S_{\rm ol}}{r^2} \sum_{i=1}^{N} \langle \mathbf{Z}^i(\hat{\mathbf{n}}^{\rm inc}, \hat{\mathbf{n}}^{\rm inc}) \rangle_\xi \mathbf{I}^{\rm inc}, \tag{30}$$

respectively, where $\langle \mathbf{Z}^i(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) \rangle_\xi$ and $\langle \mathbf{K}^i(\hat{\mathbf{n}}^{\rm inc}) \rangle_\xi$ are the phase and extinction matrices of particle i, respectively, centered at the origin of this particle and averaged over the ensemble of its microphysical states.

Second, we consider the standard energy-budget problem by (i) surrounding the multi-particle group by an imaginary sphere Σ, as shown in Fig. 6, and (ii) computing the net time-averaged flow of electromagnetic power entering the volume bounded by Σ:

$$\langle W_\Sigma \rangle_t = \langle W_\Sigma \rangle_{\mathbf{R},\xi} = -\mathrm{Re} \oiint_\Sigma \mathrm{d}^2\mathbf{r} \langle \mathbf{S}(\mathbf{r}) \rangle_{\mathbf{R},\xi} \cdot \hat{\mathbf{r}}, \tag{31}$$

where $\mathbf{S}(\mathbf{r})$ is the complex Poynting vector and "Re" stands for "the real part of". Then another fundamental result of the FOSA [39] is the following:

$$\begin{aligned} \langle W_\Sigma \rangle_{\mathbf{R},\xi} = & \sum_{i=1}^{N} \Big[\langle \mathrm{K}_{11}^i(\hat{\mathbf{n}}^{\rm inc}) \rangle_\xi I^{\rm inc} + \langle \mathrm{K}_{12}^i(\hat{\mathbf{n}}^{\rm inc}) \rangle_\xi Q^{\rm inc} + \langle \mathrm{K}_{13}^i(\hat{\mathbf{n}}^{\rm inc}) \rangle_\xi U^{\rm inc} + \langle \mathrm{K}_{14}^i(\hat{\mathbf{n}}^{\rm inc}) \rangle_\xi V^{\rm inc} \Big] \\ & - \sum_{i=1}^{N} \int_{4\pi} \mathrm{d}\hat{\mathbf{r}} \Big[\langle Z_{11}^i(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) \rangle_\xi I^{\rm inc} + \langle Z_{12}^i(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) \rangle_\xi Q^{\rm inc} \\ & + \langle Z_{13}^i(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) \rangle_\xi U^{\rm inc} + \langle Z_{14}^i(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\rm inc}) \rangle_\xi V^{\rm inc} \Big]. \end{aligned} \tag{32}$$

Equations (29)–(32) make it explicit that the FOSA exemplifies the independent scattering regime. Note however that this would not be true if the observation point was in the far zone of the entire group in the direction $\hat{\mathbf{r}} = \hat{\mathbf{n}}^{\rm inc}$. Then the exponential factor $\exp(\mathrm{i}\Delta_{ij})$ in Eq. (21) would be identically equal to one for any i and j and would survive ensemble averaging, thereby causing the so-called forward-scattering interference phenomenon [1,39,50].

10. Radiative transfer theory

The second example of the independent scattering regime is the first-principles radiative transfer theory (RTT) for sparse discrete random media [39,54]. It is based on the following fundamental assumptions:

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- Eq. (17) holds, which implies that all particles are widely separated and that the observation point resides sufficiently far from any particle;
- the observation point resides in the near zone of the volume V occupied by the particles (including the case of being inside V);
- the number of particles N in the group is very large $(N \to \infty)$;
- the N particles are moving randomly and independently of each other throughout V;
- the physical state of each particle is independent of its position and of the states and positions of all the other particles; and
- the random N-particle group is fully ergodic.

The main direct corollaries of these assumptions can be summarized as follows [39,47].

Let us first consider the reading of a polarization-sensitive WCR placed inside V and having its optical axis along the unit vector $\hat{\mathbf{q}}$ (Fig. 7). Let S be the boundary of V and S_{ill} be the part of S "directly illuminated" by a plane electromagnetic wave incident in the direction $\hat{\mathbf{n}}^{\mathrm{inc}}$. The ensemble-averaged phase and extinction matrices are defined according to

$$\langle \mathbf{Z}(\hat{\mathbf{q}},\hat{\mathbf{q}}')\rangle_{\xi} = \frac{1}{N}\sum_{i=1}^{N}\langle \mathbf{Z}^{i}(\hat{\mathbf{q}},\hat{\mathbf{q}}')\rangle_{\xi}, \quad \langle \mathbf{K}(\hat{\mathbf{q}})\rangle_{\xi} = \frac{1}{N}\sum_{i=1}^{N}\langle \mathbf{K}^{i}(\hat{\mathbf{q}})\rangle_{\xi}. \tag{33}$$

Let us further introduce an auxiliary 4-component column $\tilde{\mathbf{I}}(\mathbf{r},\hat{\mathbf{q}})$ (called the specific intensity column vector) as a function of position vector $\mathbf{r}$ and direction $\hat{\mathbf{q}}$ according to

$$\tilde{\mathbf{I}}(\mathbf{r},\hat{\mathbf{q}}) = \delta(\hat{\mathbf{n}}^{\mathrm{inc}} - \hat{\mathbf{q}})\mathbf{I}_{\mathrm{c}}(\mathbf{r}) + \tilde{\mathbf{I}}_{\mathrm{d}}(\mathbf{r},\hat{\mathbf{q}}), \tag{34}$$

where the "coherent" part is the solution of the boundary-value problem

$$\hat{\mathbf{n}}^{\mathrm{inc}} \cdot \nabla \mathbf{I}_{\mathrm{c}}(\mathbf{r}) = -n_0 \langle \mathbf{K}(\hat{\mathbf{n}}^{\mathrm{inc}})\rangle_{\xi} \mathbf{I}_{\mathrm{c}}(\mathbf{r}), \quad \mathbf{I}_{\mathrm{c}}(\mathbf{r})\big|_{\mathbf{r}\in S_{\mathrm{ill}}} = \mathbf{I}^{\mathrm{inc}}, \tag{35}$$

and the "diffuse" part is the solution of the boundary-value problem

$$\begin{aligned} \hat{\mathbf{q}} \cdot \nabla \tilde{\mathbf{I}}_{\mathrm{d}}(\mathbf{r},\hat{\mathbf{q}}) &= -n_0 \langle \mathbf{K}(\hat{\mathbf{q}})\rangle_{\xi} \tilde{\mathbf{I}}_{\mathrm{d}}(\mathbf{r},\hat{\mathbf{q}}) + n_0 \int_{4\pi} \mathrm{d}\hat{\mathbf{q}}' \langle \mathbf{Z}(\hat{\mathbf{q}},\hat{\mathbf{q}}')\rangle_{\xi} \tilde{\mathbf{I}}_{\mathrm{d}}(\mathbf{r},\hat{\mathbf{q}}') \\ &+ n_0 \langle \mathbf{Z}(\hat{\mathbf{q}},\hat{\mathbf{n}}^{\mathrm{inc}})\rangle_{\xi} \mathbf{I}_{\mathrm{c}}(\mathbf{r}), \quad \tilde{\mathbf{I}}_{\mathrm{d}}(\mathbf{r},\hat{\mathbf{q}}_{\leftarrow})\big|_{\mathbf{r}\in S} = \mathbf{0}. \end{aligned} \tag{36}$$

In these formulas, $n_0 = N/V$ is the average number of particles per unit volume; $\hat{\mathbf{q}}_{\leftarrow}$ is any unit vector directed *into* the volume V; $\delta(\hat{\mathbf{q}})$ is the solid-angle delta function; and $\mathbf{0}$ is a zero 4-component column.

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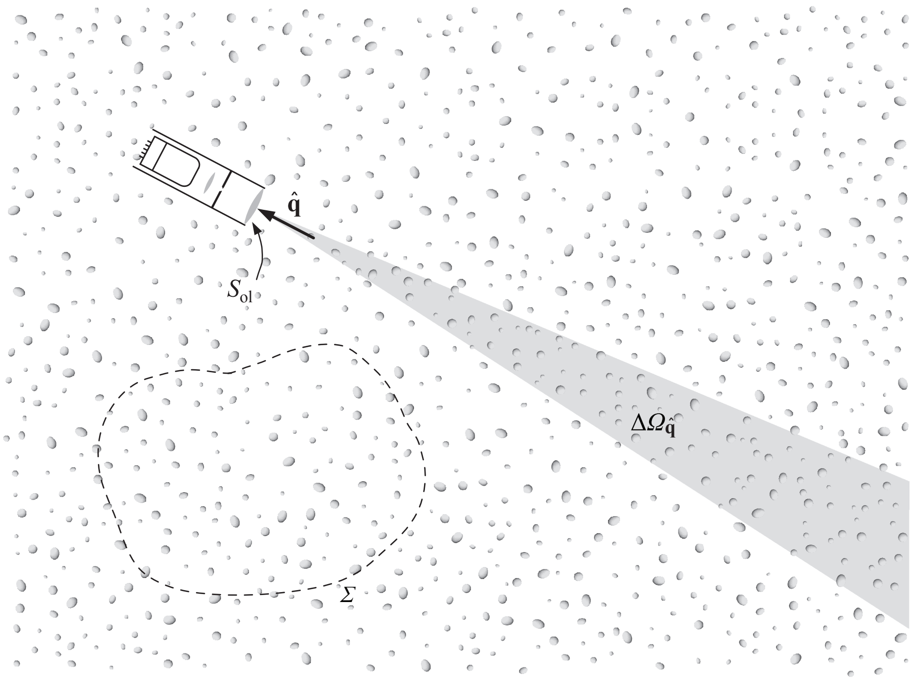

Fig. 7. Sparse discrete random medium. The sizes of the particles and the WCR are exaggerated for demonstration purposes.

Then the temporal average of the polarized reading of the WCR in Fig. 7 per unit time is equal to the corresponding ensemble average and can be computed according to

$$\langle \textbf{Signal}(\mathbf{r},\hat{\mathbf{q}})\rangle_{\mathbf{R},\xi} = S_{\text{ol}}\int_{\Delta\Omega_{\hat{q}}} \mathrm{d}\hat{\mathbf{q}}'\tilde{\mathbf{I}}(\mathbf{r},\hat{\mathbf{q}}') \approx \begin{cases} S_{\text{ol}}\mathbf{I}_{\text{c}}(\mathbf{r}) + S_{\text{ol}}\Delta\Omega\tilde{\mathbf{I}}_{\text{d}}(\mathbf{r},\hat{\mathbf{n}}^{\text{inc}}) & \text{if } \hat{\mathbf{q}} = \hat{\mathbf{n}}^{\text{inc}}, \\ S_{\text{ol}}\Delta\Omega\tilde{\mathbf{I}}_{\text{d}}(\mathbf{r},\hat{\mathbf{q}}) & \text{if } \hat{\mathbf{q}} \neq \hat{\mathbf{n}}^{\text{inc}}, \end{cases} \tag{37}$$

where $\Delta\Omega$ is the WCR's acceptance solid angle. Note that Eq. (37) is essentially based on the specific functionality of the WCR as a narrow-angle filter of wave-propagation directions rather than spurious "energy propagation directions" [39,51,54]. As a consequence of this functionality, the WCR in Fig. 7 reacts only to those scattering sequences in the expansion (17) that have their last particles residing in the conical acceptance volume shown by gray color.

The same auxiliary quantity $\tilde{\mathbf{I}}(\mathbf{r},\hat{\mathbf{q}})$ can be used to evaluate the energy balance of a finite volume of discrete random medium bounded by the closed surface Σ (Fig. 7). Again, the net time-averaged flow of electromagnetic power entering the volume bounded by Σ is

$$\langle W_\Sigma\rangle_t = \langle W_\Sigma\rangle_{\mathbf{R},\xi} = -\mathrm{Re}\oiint_\Sigma \mathrm{d}^2\mathbf{r}\langle \mathbf{S}(\mathbf{r})\rangle_{\mathbf{R},\xi}\cdot\hat{\mathbf{n}}(\mathbf{r}), \tag{38}$$

where $\hat{\mathbf{n}}(\mathbf{r})$ is the unit vector in the direction of the local outward normal to Σ. Then

$$\mathrm{Re}\langle \mathbf{S}(\mathbf{r})\rangle_{\mathbf{R},\xi} = \int_{4\pi}\mathrm{d}\hat{\mathbf{q}}\,\hat{\mathbf{q}}\tilde{I}(\mathbf{r},\hat{\mathbf{q}}), \tag{39}$$

where $\tilde{I}(\mathbf{r},\hat{\mathbf{q}})$, called the specific intensity, is the first element of $\tilde{\mathbf{I}}(\mathbf{r},\hat{\mathbf{q}})$.

The inspection of Eqs. (33)–(39) confirms that the assumptions listed in the beginning of this section result in the independent scattering regime.

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An interesting interference phenomenon not captured by Eq. (37) is the so-called coherent backscattering effect. It can be observed if the WCR is located sufficiently far from the particulate medium (ideally in its far zone) and has its optical axis along the backscattering direction (see, e.g., Chapters 18 and 21 of Ref. [39], Chapter 14 of Ref. [47], and references therein). Coherent backscattering exemplifies the dependent scattering regime.

11. Discussion and conclusions

We have pointed out in Section 3 that in the framework of frequency-domain electromagnetic scattering, the entire multi-particle group acts as a unified, albeit morphologically complex, scattering object. We therefore started our analysis by formulating the basic scattering problem for the entire multi-particle object and then traced the FOSA and the RTT as direct consequences of the first-principles VIE formalism coupled with specific micro- and macrophysical assumptions about the object (sparsity, the limit of $N \to 0$ or $N \to \infty$, randomness, ergodicity, etc.) and the deliberate consideration of an observation point located in the near zone of the object. We have found that as a result of an explicit first-principles derivation, both the FOSA and the RTT are formulated in terms of basic single-particle far-field observables (i.e., the phase and extinction matrices) and hence are manifestations of the independent scattering regime.

The basic *a priori* premise in many previous studies has been the belief that if the individual far-field optical observables of each constituent particle are known then all scattering properties of the entire multi-particle group can somehow be constructed from those of the constituent particles using vague “physical obviousness” as the main argument. Accordingly, the traditional *ad hoc* methodology has been to proceed in the direction exactly opposite to that of our first-principles approach:

- by first computing far-field optical observables of each particle in total isolation from all the other particles;
- then postulating that widely separated particles forming a particle group can be considered “independently scattering” and characterized individually by the previously determined extinction and phase matrices;
- then considering “incoherent single scattering” by the independently scattering particles occupying an imaginary small (“elementary”) volume element;
- then postulating the FOSA for a small volume element and deriving (essentially postulating) the phenomenological radiative transfer equation by considering “incoherent multiple scattering” by small volume elements serving as building blocks of the particulate medium; and finally
- by speculating how the single-scattering properties of the individual particles and of the small volume elements can change as a consequence of hypothetical “packing density” effects.

Again, this questionable approach is based on the lack of recognition that from the fundamental perspective of electromagnetics, the entire particulate medium is a unified scattering object and must be treated as such from the outset.

Our first-principles analysis appears to imply that the FOSA and RTT may be the only notable manifestations of the independent scattering regime, all other cases of electromagnetic scattering by particulate media belonging to the category of dependent scattering. If so, the terms “independent scattering” and “dependent scattering” have limited heuristic value, and their use can probably be avoided altogether by referring directly to the FOSA and the RTT as opposed to any other scattering scenario. Then the bulleted lists of assumptions in the beginning of Sections 9 and 10, respectively, can serve as operational definitions of the independent scattering regime in the cases of a sufficiently small and a very large *N*.

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It should be recognized that the above analysis is expressly based on the frequency-domain theory of elastic electromagnetic scattering. Given the widespread use of pulsed light sources, some may wonder how the concept of independent scattering might look with a femtosecond pulse impinging on a collection of particles. One could imagine a case where the particles are close enough together as to certainly be in the dependent scattering regime for a monochromatic incident wave, yet perhaps that same collection becomes in some sense "independent" for an incident pulse that is short enough. Of course, in such cases even the theory of electromagnetic scattering by a single particle may need a major modification [55]. Another aspect that needs a first-principle analysis is thermal emission by sufficiently hot particles; this is an issue frequently encountered in heat transfer applications (see, e.g., Refs. [2,25,28,56,57] and numerous references therein).

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