



Dependent light scattering in dense heterogeneous media

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Abstract

We have developed a new tool for the calculation of multiple-dependent scattering in dense heterogeneous media avoiding convergence difficulties. We demonstrate with various examples the influence of the multiple-dependent scattering on the scattering cross sections and the diffuse reflectance (or transmittance) as calculated by using an extended radiative transfer model. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Recent years have seen a renewed interest in the multiple scattering of light by heterogeneous media, essentially motivated by needs arising in applications. The principal interest is the electromagnetic wave scattering by dense media containing a large number of strongly scattering inclusions (size comparable to or larger than the wavelength). The related applications are in the domains of paints, papers, cosmetics, astrophysics, biomedical imaging, etc.

In most applications, one requires a complete treatment of both multiple and dependent scatterings. More than a century ago, the question of electromagnetic single scattering by spheres was solved by Gustav Mie. The question of multiple scattering has since been repeatedly addressed, but never satisfactorily solved in practice, because of the strong interplay between its analytical and numerical aspects, and the complexity of the latter, due to the unavoidable field expansion in terms of a theoretically infinite set of spherical partial waves involved in different steps of the treatment.

In recent years, Borghese [1], Mackowski [2] and Xu [3] have made decisive advances towards the practical solution of this question. Nevertheless, these treatments are unable to account, by themselves, for the experi-

mental properties of random media with well-defined boundaries, for instance thin or thick films with parallel interfaces. These properties are characterized by the measurements of the scattered, transmitted, or reflected energy flux. The radiative transfer theory in the form of 2, 4 or N-flux models is undoubtedly the most efficient tool for solving this problem. However, its treatment of multiple scattering fails in dense media where the interactions between scatterers are strong (dependent scattering).

These considerations justify the developments we propose in this paper for the treatment of the multiple-dependent scattering in dense random media. Our proposal is twofold:

- (1) at the microscopic scale, we perform an exact local electromagnetic calculation of the field scattered by an assembly of a limited number of strongly interacting spheres. Our calculation is based on a recursive T -matrix algorithm that we modified in order to avoid the convergence problems inherent to this formulation. The cross sections are then evaluated. This kind of treatment allows the calculation of the scattering by a cluster of any shape, approximated by an appropriate arrangement of spheres.
- (2) at the macroscopic scale, we calculate the reflected and transmitted flux scattered by a parallel slab of such a material. We assume that the local exact calculation has been performed up to a scale corresponding to a coherence length that we define as the length below which coherent and dependent scatterings have to be taken into account. Beyond this

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scale, we apply a four flux model derived from the radiative transfer equation which uses the output of the local calculation.

2. Multiple-scattering calculation

2.1. Scattering by one sphere

According to Mie theory, a time harmonic electric field (\mathbf{E}_0) impinging on a dielectric sphere and the corresponding scattered field (\mathbf{E}_{scat}) can be expanded in terms of spherical vector wave functions $\mathbf{M}_{mn}^{(i=1,3)}(kr, \theta, \phi)$ and $\mathbf{N}_{mn}^{(i=1,3)}(kr, \theta, \phi)$. The T -matrix converts the incident field expansion coefficients a_{mn} into expansion coefficients f_{mn} of the radiated scattering field via the relation

$$\begin{pmatrix} f_{1\ mn} \\ f_{2\ mn} \end{pmatrix} = \begin{pmatrix} T_n^{11} & 0 \\ 0 & T_n^{22} \end{pmatrix} \begin{pmatrix} a_{1\ mn} \\ a_{2\ mn} \end{pmatrix}. \quad (1)$$

2.2. Scattering by an assembly of dielectric spheres

We now consider an assembly of N spheres. Assuming multiple scattering to occur, the total field impinging upon the i th scatterer (excitation field $\mathbf{E}_{\text{exc}}^{(i)}$), is the sum of the contributions of the field from the original source, \mathbf{E}_0 , and the scattered fields, $\mathbf{E}_{\text{scat}}^{(j)}$, from all other scatterers. Using the T -matrix formulation of the single-scattering formalism, one can define N coupled linear equations whose unknowns are the expansion of the scattered field of each individual sphere. $i = 1, \dots, N$

$$\mathbf{E}_{\text{scat}}^{(i)} = \bar{\mathbf{T}}_1^{(i)} \cdot \left[\mathbf{E}_0 + \sum_{\substack{j=1 \\ j \neq i}}^N \mathbf{E}_{\text{scat}}^{(j)} \right]. \quad (2)$$

In the above equation, the incident field is expanded in the principal coordinate system, whereas the scattered field from each sphere is expressed as outgoing waves centered, respectively, on the corresponding scatterer. In order to obtain equations in terms of the expansion coefficients, one must invoke the translation–addition theorem for the spherical vector wave functions, in order to re-reference all fields in the i th scatterer coordinate system:

$$\mathbf{f}_N^{(i)} = \bar{\mathbf{T}}_1^{(i)} \left[\bar{\beta}(i, 0) \cdot \mathbf{a}^0 + \sum_{\substack{j=1 \\ j \neq i}}^N \bar{\alpha}(i, j) \cdot \mathbf{f}_N^{(j)} \right], \quad i = 1, \dots, N, \quad (3)$$

where $\bar{\beta}(i, 0)$ and $\bar{\alpha}(i, j)$ are, respectively, the regular and irregular translation–addition matrices. In order to solve the linear system, it is convenient to first introduce the N -scatterer T -matrix of the i th scatterer, $\bar{\mathbf{T}}_N^{(i)}$, which includes all the information about multiple scattering effects due to the presence of N scatterers. It directly links

the incident field \mathbf{E}_0 to the scattered field of the i th scatterer. The single analytical expression of the total field scattered by the assembly is obtained via the superposition principle and requires that individual scattered fields be translated to a common basis.

We have experienced that this kind of formalism leads to severe convergence problems related to the series expansions in the regular translation matrix. One of our contributions in this domain has been to introduce a hybrid technique for multiple-scattering computations of assemblies of spheres. We modified an efficient recursive T -matrix algorithm of the type introduced by Chew [4] (Eqs. (4a) and (4b)) in order to obtain recursion relations free of the troublesome regular translation matrices (Eq. (4c)). An important aspect of our calculations is that they keep all the information concerning the fields in the interior of the assembly which is important when evaluating the modifications of individual particle scattering due to coherence effects. When calculating amplitude scattering matrices and differential cross sections, we have put outgoing wave phase shifts of the type introduced by Xu [3] (Eq. (5a)) on an equal footing with incoming wave phase shifts (Eq. (5b)). Total cross-sections of the cluster (Eq. (5c)) are deduced from formulations of the type introduced by Borghese [1] and Mackowski [2].

$$\begin{aligned} \bar{\mathbf{T}}_{N+1}^{(N+1)} = & \left[\bar{\mathbf{I}} - \bar{\mathbf{T}}_1^{(N+1)} \sum_{i=1}^n \bar{\alpha}(N+1, i) \cdot \bar{\mathbf{T}}_N^{(i)} \cdot \bar{\alpha}(i, N+1) \right]^{-1} \\ & \cdot \bar{\mathbf{T}}_1^{(N+1)} \cdot \left[\bar{\mathbf{I}} + \sum_{i=1}^n \bar{\alpha}(N+1, i) \cdot \bar{\mathbf{T}}_N^{(i)} \right. \\ & \left. \cdot \bar{\beta}(i, N+1) \right], \end{aligned} \quad (4a)$$

$$\bar{\mathbf{T}}_{N+1}^{(i)} = \bar{\mathbf{T}}_N^{(i)} \cdot [\bar{\mathbf{I}} + \bar{\alpha}(i, N+1) \cdot \bar{\mathbf{T}}_{N+1}^{(N+1)} \cdot \bar{\beta}(N+1, i)], \quad (4b)$$

$$\bar{\mathbf{T}}_{N+1}^{(i)} = \sum_{k=1}^N \bar{\mathbf{T}}_N^{(i,k)} \cdot \bar{\beta}(k, i), \quad (4c)$$

$$\mathbf{f}_N = \sum_{i=1}^N \mathbf{f}_N^{(i)} e^{-i\mathbf{k}_{\text{scat}} \cdot \mathbf{x}_i}, \quad (5a)$$

$$\mathbf{f}_N^{(i)} = e^{i\mathbf{k}_{\text{inc}} \cdot \mathbf{x}_i} \bar{\mathbf{T}}_N^{(i)} \cdot \mathbf{a}_0, \quad (5b)$$

$$\bar{\mathbf{T}}_N = \sum_{i=1}^N e^{-i\mathbf{k}_{\text{scat}} \cdot \mathbf{x}_i} \bar{\mathbf{T}}_N^{(i)} e^{i\mathbf{k}_{\text{inc}} \cdot \mathbf{x}_i}. \quad (5c)$$

3. Modeling physical problems

The multiple-scattering technique described in the last section is a powerful tool which can be applied to many physical problems. We show here two applications: (i) the evaluation of a coherence length related to the transition from independent to dependent scattering behaviors; (2) the modeling of scattering by particles of arbitrary shape represented by an appropriate arrangement of spheres.

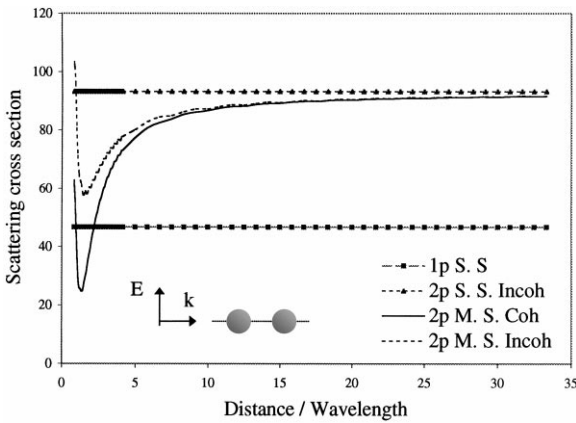


Fig. 1. Scattering cross-section by a system of two identical spheres. Relative index of refraction 1.5, size parameter 5.2: (1p SS) one sphere (simple scattering); (2p SS) two spheres (incoherent simple scattering); (2p MS) two spheres (multiple scattering); (2p MS) two spheres (incoherent multiple scattering).

3.1. Coherence length in assemblies of two and nine interacting spheres

We present in Fig. 1 the calculation of the variation of the total scattering cross section of two identical spheres as a function of their separation. The relative refractive index of the spheres is $N = 1.5$ and their size parameter (circumference to wavelength ratio) is 5.2. The incident wave vector is parallel to the symmetry axis. We have calculated the scattering cross section of a single isolated sphere and of the two sphere system which is compared to the total scattering cross section of the system with multiple-scattering effects only and multiple-scattering plus coherence effects. Three different domains show up in the scattering cross section curve related to different characteristic coherence lengths. At large separations multiple scattering and coherence effects are negligible. In an intermediate region, the single-scattering approximation is no longer valid and the multiple-scattering process has to be taken into account while the coherence effects are still negligible. In the region of small separation, coherence effects between scattered fields by each sphere appear to be quite important.

The above system is now extended to a system of nine identical spheres placed on a cubic centered lattice. Their relative index is still 1.5, while their size parameter is 5.0 and the incident wave vector is normal to a face of the cube. In Fig. 2 are represented: the scattering factor s (scattering cross section C_{scat} multiplied by the number of sphere per unit volume N_p) of the whole system and of the central sphere as a function of the volume fraction p , in the cases of isolated and interacting spheres. One can observe that the transition between independent and

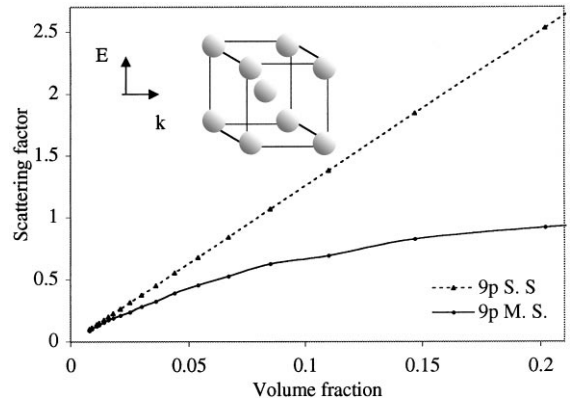


Fig. 2. Scattering factor of a system composed of nine identical spheres. Relative index of refraction 1.5, size parameter 5.0: (9p SS): nine spheres (simple scattering); (9p MS) nine spheres (multiple scattering).

dependent behaviors at a given volume fraction is well accounted for by our scattering model.

3.2. Scattering by an aggregate of spheres

We present in Fig. 3 the scattering cross section in 2D and 3D of an aggregate of 13 spheres of index of refraction 1.59 and size parameter 11, compared to the same quantity for a single sphere of the same index and a size parameter 35, corresponding to the same overall radius as calculated by our technique. One observes a decrease of one order of magnitude of the scattering, the occurrence of anisotropy, and the curvature of the plane sections of the scattering lobes.

4. Extended radiative transfer model

We propose to extend the four flux radiative transfer model [5] by introducing, at a local scale, multiple scattering and coherent effects via the electromagnetic theory presented in Section 2. An effective scattering cross section is associated with each particle which tends towards the scattering cross section of an isolated sphere in dilute media. Fig. 4 presents a comparison of the predictions of simple and extended four flux models concerning the diffuse reflectance of different samples: spheres of radius $0.25 \mu\text{m}$, volume fractions 0.012 (Fig. 4a) and 0.132 (Fig. 4b) and sample thicknesses 1.5 and $10 \mu\text{m}$. The incident flux on the heterogeneous medium is totally collimated. A significant difference in the predictions of the two models already appears, in this example, at volume fractions of a few percent. This preliminary calculation only takes into account bi-sphere interactions.

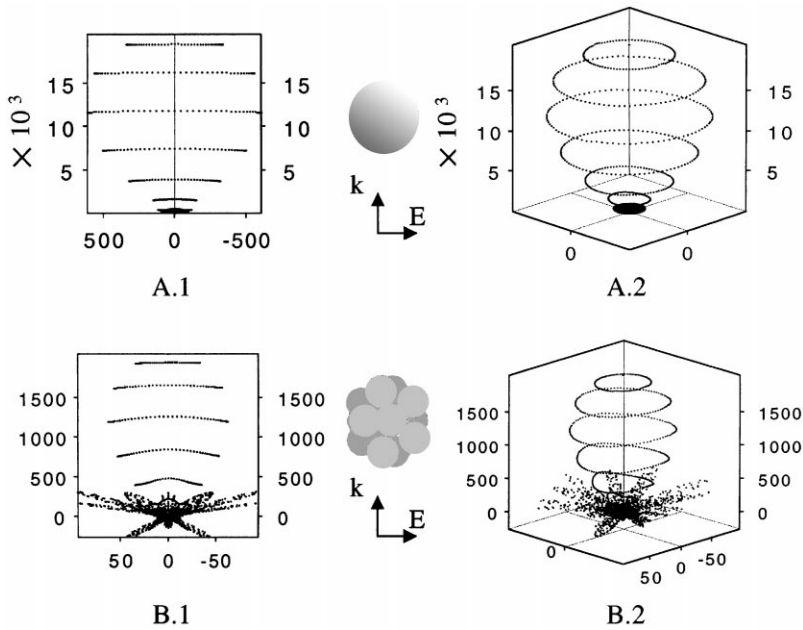


Fig. 3. Differential scattering 2D and 3D patterns: (A) Single sphere of relative index of refraction 1.59, size parameter 35; (B) cluster of 13 identical spheres (with the same overall radius) of relative index of refraction 1.59, size parameter 11. Subscript 1 and 2 are for $\theta = 0^\circ$, $\phi = 90^\circ$ and $\theta = 22.5^\circ$, $\phi = 135^\circ$ views.

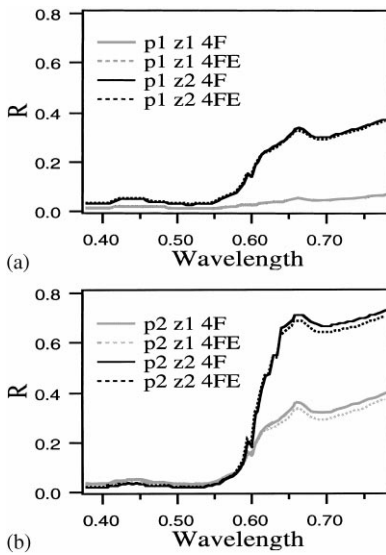


Fig. 4. Comparison of the predictions of simple (4F) and extended (4FE) four flux models for the diffuse reflectance of spheres of radius $0.25 \mu\text{m}$, volume fractions 0.012 (Fig. 4a) and 0.132 (Fig. 4b) and sample thicknesses 1.5 and $10 \mu\text{m}$.

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