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Light Scattering From Irregular Tetrahedral Aggregates

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ARL-TR-1726

February 1999

Army Research Laboratory

Adelphi, MD 20783-1197

ARL-TR-1726 **February 1999**

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Abstract

We examine the light scattering from spheres aggregated into tetrahedral structures and the changes that occur when the internal stucture of the particle system is modified by filling the central portion of the tetrahedron with an index-matching solution. Several effects are noticed in the scattering signals. The scattering phase function of aggregates is quite sensitive to changes in the internal structure within the aggregate. The positions and relative intensities of the maxima and minima of an aggregate tend to have a stronger dependence on orientation and internal structure than those of a sphere containing an inclusion. As the area of the interfaces increases, the backscatter intensity increases, resulting in a decrease in the asymmetry parameter. Specular reflections also occur because of the constructive interference of rays reflecting off the different lattice sites.

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1. Introduction

Mie theory predicts the electromagnetic fields scattered by a homogeneous sphere that is illuminated by a plane wave. Because of its relative simplicity, a homogeneous sphere is often used to model the light scattered from irregular particle systems, despite that the light scattered from these spheres is often significantly different than that scattered by other types of particles [1–4]. One distinguishing characteristic of the light scattered from spheres is the presence of morphology dependent resonances (MDRs) [5]. When the spherical symmetry is broken, either through the inclusion of surface perturbations or volume irregularities, these resonant peaks broaden and diminish in amplitude [6,7]. Another characteristic of spheres is that they generally scatter a larger proportion of light into the forward direction than they scatter irregular particles; i.e., their asymmetry parameters may be significantly larger than the particles for which they are being used to model [1–4]. The symmetry of spherical particles provides an additional identifying feature, since the total intensity of their scattered light has no azimuthal dependence. Although the scattering phase functions of nonspherical particles are often overwhelming because they have both a θ and ϕ dependence, this additional scattering information can be used to characterize the system.

One type of nonspherical particle with interesting optical properties is an aggregate made up of many spherules in contact. Carbon (soot) particles form loose fractal-like aggregates having Rayleigh-sized spherical components [8]. Since these particles play a role in the earth radiation budget, their scattering properties are of especial interest [9]. Other aggregates of interest are clusters of biowarfare agent spores. When a liquid solution containing such spores is aerosolized and the liquid evaporates, compact and nearly spherical clusters of these spores remain. Early warning systems depend on rapid identification of these particles, and elastic and fluorescent light scattering may play a crucial role in the development of such systems. The motivation for our studies is the characterization of this latter type of particle through elastic light scattering [10].

One of the driving questions in our research is what effect does the internal structure of a particle system have on its scattered field? In our studies of spore aggregates (and for many other types of aggregates), no prior information exits on their internal structure: either the orientation of the individual spores with respect to each other, the addition of contaminants in the lattice, or whether a liquid core fills the gaps between the spores. The internal structure may have a drastic effect on the accuracy of modeling efforts.

2. Results

We obtained the scattering results shown in this report by using a finite-difference time domain (FDTD) algorithm with a perfectly matched absorbing boundary condition, which is a direct solution of Maxwell's time-dependent curl equations [11–15]. This method treats the scattering and absorption of the particle as an initial-value problem. It should be noted that this scheme requires a descritization of the particle system, so it is relatively computer intensive compared with multipole techniques that are extremely efficient at calculating the scatter from aggregates of spheres [16–27]. However, in our studies, we are interested in the dependence of the scatter on the internal structure of the particle, and in some cases, the aggregate components could not be described as spheres. Multipole techniques are incapable of handling such systems. To verify the validity of the calculations, we compare our results with those calculated using multipole techniques. Figure 1 shows a comparison of the scatter from a pair of spheres in contact, illuminated end-on, calculated using the multipole method and the FDTD algorithm with a mesh resolution of $\lambda/30$. The refractive index, $m = 1.53 + 0.001i$, approximates that of biological spores [28,29]. Only a small discrepancy exists between the calculated results of the two models.

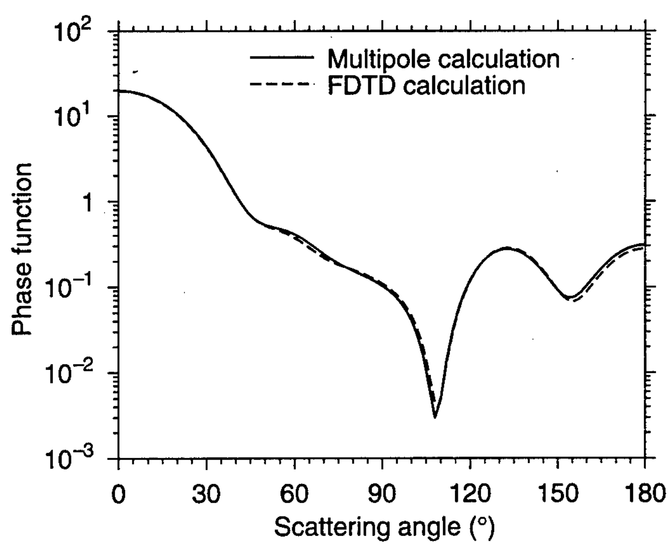


Figure 1. Light scattering phase function from a pair of $x = \pi$ $(a = \lambda/2)$, $m = 1.53 + 0.001i$ spheres in contact, illuminated end-on, calculated using multipole method (solid line) and FDTD algorithm with a mesh resolution of $\lambda/30$ (dashed line).

One system that can provide insight into the effect of internal structure on the scattering from an aggregate system is that of a sphere containing a spherical inclusion [30–35]. In this system, the amount of internal structure can be varied by changing the size and optical properties of the inclusion. Figure 2 shows the light scattering Mueller matrix for two spherical particles, one of which has a large absorbing inclusion. The gross scattering properties change drastically when the inclusion is present: the absorption efficiency increases from Q_{abs} = 0.0333 with no inclusion present to Q_{abs} = 0.298 when the inclusion is present, and the scattering efficiency decreases from Q_{sca} = 2.021 with no inclusion present to Q_{sca} = 2.0165 when the inclusion is present. Interestingly, compared with these efficiencies, the amplitude of the phase function (proportional to matrix element S_{11}) is only slightly affected by the presence of the inclusion. The positions of the maxima and minima and the relative heights change only slightly. The polarization state of the scattered light is more sensitive to the presence of the inclusion than to the total intensity. In summary, even though the inclusion represents a substantial volume of the particle system and changes the absorption efficiency of the particle system significantly, it has little effect on the scattering phase function. (Note that these conclusions may not apply when the inclusion is placed in the region very near the edge of the sphere.) As we shall see, this is not the case for aggregates.

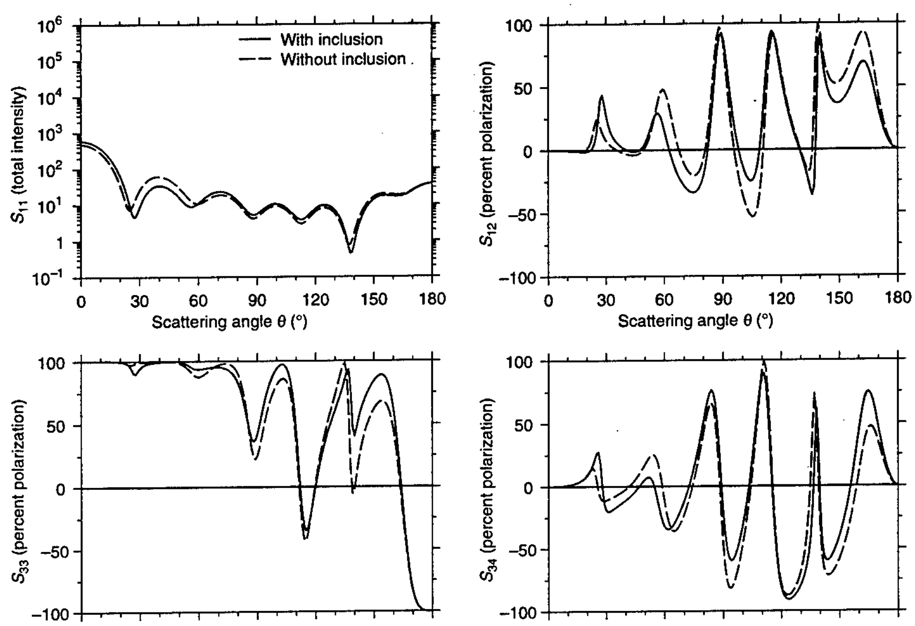


Figure 2. Light scattering Mueller matrix as a function of scattering angle of an $m_{host} = 1.53 + 0.001i$, $a_{host} = \lambda$ sphere containing an $m_{inc} = 1.94 + 0.66i$, $a_{inc} = a_{host}/4$ spherical inclusion offset a distance $d = a_{host}/4$ in forward-scattering direction (solid line). Also shown are matrix elements for sphere without inclusion (dashed line).

One of the simplest approximations to an aggregation of spores is four spherules in a tetrahedral orientation as shown in figure 3. This system is digitized and used in the FDTD algorithm to calculate the resulting scattering phase function. The easiest method of examining the effect of the internal structure of the scattering system is to remove it by filling the central portion of the tetrahedron with a refractive-index-matching solution. We performed this by setting the refractive index in the region $r < \lambda/2$ to $m = 1.53 + 0.001i$ (equivalent to the refractive index of the other spherules). This is equivalent to filling the air gaps between the spherules with a cell-like substance to a distance equal to the radius of the spherules. The scattering phase function averaged over ϕ is shown in figure 4 for two incident orientations: Figure 4a shows the case of a plane wave incident upon the particle from the bottom (angle of incidence is 0°), and figure 4b shows the case of a plane wave incident upon the particle from the top (angle of incidence is 180°). There are several results that can be gleaned from these figures.

We expect and see from figure 4 that an extremely strong dependence is on the particle orientation. Indeed, the phase functions shown in figure 4a are quite different from those of figure 4b. From our calculations of the phase functions of spheres shown in figure 2, we might also expect that the internal structure does not have a large effect on the phase function, but this is not the case. Unlike the minor changes that occur in the phase function when the internal structure of a spherical system is modified, changes in the phase function of the aggregate have a much stronger dependence on its internal structure, particularly for 180° incidence. The positions of the maxima and minima shift (apparent in both figs. 4a and

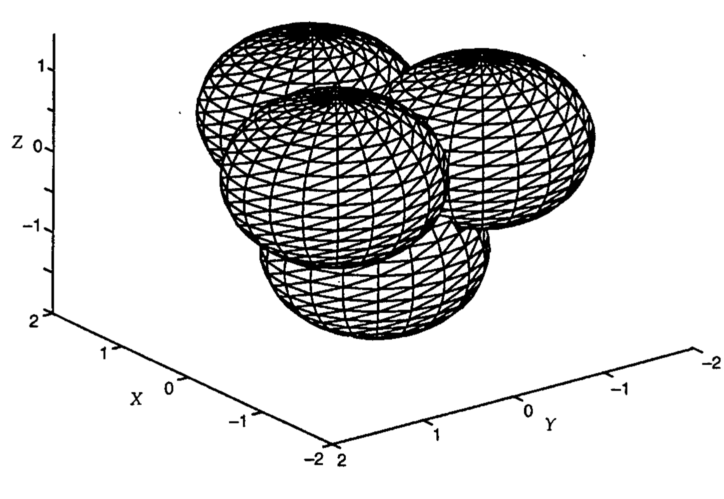
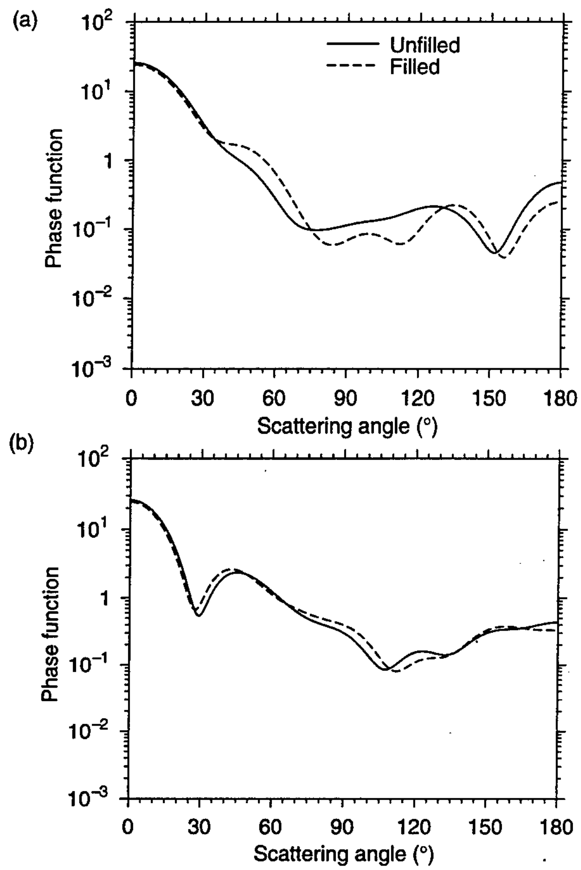


Figure 3. Diagram of tetrahedral scattering system used in our simulations. Four $a = \lambda/2$, $m = 1.53 + 0.001i$ spheres are in contact. To study effect of internal structure, we set refractive index in region $r < \lambda/2$ to $m = 1.53 + 0.001i$. Angle of incidence of illuminating plane wave is measured from positive z axis.

Figure 4. Azimuthally averaged scattering phase functions from system illustrated in figure 3: (a) plane wave is incident upon particle from bottom (angle of incidence is 0°), and (b) plane wave is incident upon particle from top (angle of incidence is 180°).

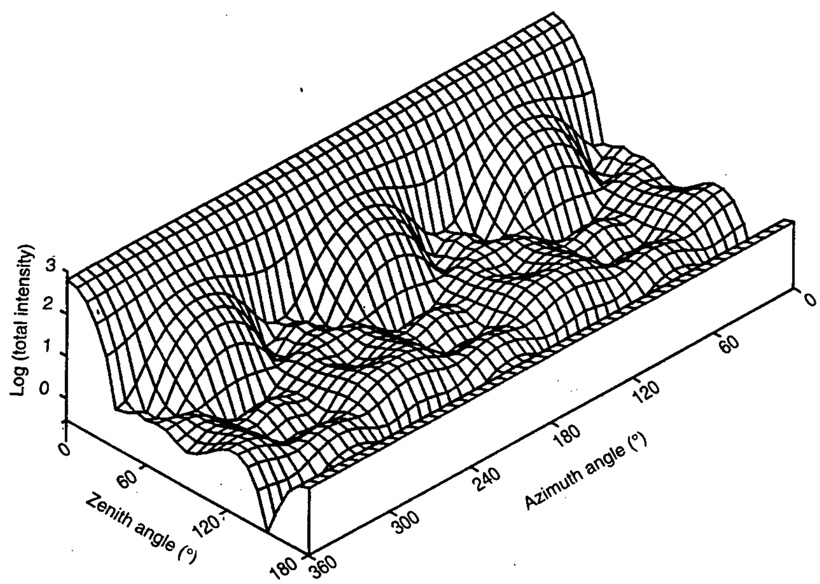


4b), and their relative amplitudes also change significantly. In addition, new maxima and minima occur in the phase function where there was none, or only minor perturbations existed previously (apparent in fig. 4a). These changes are quite significant despite that the phase function is averaged over the azimuthal angle ϕ. From our simulations, the structure of the phase function of a spherical system is much harder to perturb through the introduction of volume perturbations within the particle than the structure of an aggregate system.

Another effect of changing the internal structure of the particle system is a change in the backscatter intensity. Enhanced backscatter is the result of constructive interference of reciprocal light rays interacting with multiple sites in a particle system [36]. Particles having more surface area are expected to have a more dominant backscatter peak, since light rays striking the particle system have more paths to reach the backscatter direction after multiple reflections. We see this is the case in our simulations shown in figure 4. The unfilled tetrahedral structures have more surface area and consequently a higher backscatter peak. Enhanced backscatter affects the asymmetry parameter, a vital input in many global climate models. For both incident angles illustrated in figure 4, the asymmetry parameter decreases when the enhanced backscatter increases. When the incident angle is 0°, the asymmetry parameter decreases from $g = 0.788$ when the structure is unfilled to $g = 0.787$ when the structure is filled (a decrease of $\sim$ 0.1 percent). When the incident angle is 180° the asymmetry parameter decreases from $g = 0.643$ when the structure is unfilled to $g = 0.621$ when the structure is filled (a decrease of $\sim$ 3.5 percent). These results are relevant to scattering in the atmosphere. Ice crystals in cirrus clouds, for instance, tend to form irregular aggregates. Because cirrus clouds cover a significant portion of the earth, they influence the climate through their effects on the radiation budget. In many spectral regions, scattering algorithms tend to overestimate the asymmetry parameters of these crystals [4]. One reason for this is that these algorithms do not account for the effects of aggregation on the asymmetry parameter of the ice crystals.

One other effect of the scattering from aggregate systems is constructive interference that occurs between rays scattered from the different lattice sites. The planes that intersect lattice sites can be thought of as mirrors that reflect the incident beam. One lattice plane of the tetrahedral structure shown in figure 3 is parallel to the x-y plane. As a result, backscatter rays from the spherules located on these lattice sites also contribute to an enhancement of the backscatter peak. Three other planes are oriented at approximately 20° with respect to the z axis. We expect to see an enhancement at three locations of the azimuthal angle $\phi = 60°$, 180°, and 300° when the zenith angle is approximately $\theta = 40°$. Figure 5 shows the light scattering total intensity as a function of the zenith and azimuthal angles for the unfilled tetrahedral structure shown in figure 3 for an incident angle of 0°. Evident in this figure are distinct increases in the intensity at these locations. It is interesting to note that the minima that occur at $\phi = 0°$, 120°, and 240°, almost completely cancel the effects of these maxima, so that this structure is not apparent when the phase function is averaged over the azimuthal angle ϕ (shown in fig. 4a).

Figure 5. Angular dependence of total scattered intensity of system illustrated in figure 3 when plane wave is incident upon particle from bottom (angle of incidence is 0°).



3. Conclusion

In our simulations of irregular aggregates, we found that the internal structure (i.e., filling the central portion of the tetrahedron with a refractive-index-matching solution) has an extremely strong impact on the scattering phase function. Changes made to the internal structure of a spherical system (for instance, adding a large inclusion) may not significantly alter the positions of the maxima and minima or their relative amplitudes in the scattering phase function. This information is quite important in inversion schemes, since it allows the size and optical properties of the host to be found. Internal structure in the aggregate shifts these positions and can even add new maxima and minima, making sizing difficult. The internal structure also affects the amount of enhanced backscatter, thereby affecting the asymmetry parameter. However, the additional information provided in the azimuthally dependent phase function can provide information on the lattice structure of the aggregate.

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Adelphi MD 20783-1197

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY *(Leave blank)*	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	February 1999	Final 1 Oct 97 to 1 Jun 98

4. TITLE AND SUBTITLE Light Scattering From Irregular Tetrahedral Aggregates

5. FUNDING NUMBERS
DA PR: B53A
PE: 61102

6. AUTHOR(S) Gorden Videen (ARL), Wenbo Sun, Qiang Fu (Atmospheric Sciences Program, Dept of Oceanography, Dalhousie University, Nova Scotia)

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
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2800 Powder Mill Road
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email: gvideen@arl.mil

8. PERFORMING ORGANIZATION REPORT NUMBER
ARL-TR-1726

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
U.S. Army Research Laboratory
2800 Powder Mill Road
Adelphi, MD 20783-1197

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES
ARL PR: 7FEJ70
AMS code: 61110253A11

12a. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

We examine the light scattering from spheres aggregated into tetrahedral structures and the changes that occur when the internal structure of the system is modified by filling the central portion of the tetrahedron with an index-matching solution. Several effects are noticed in the scattering signals. The scattering phase function of aggregates is quite sensitive to changes in the internal structure within the aggregate. The positions and relative intensities of the maxima and minima of an aggregate tend to have a stronger dependence on orientation and internal structure than those of a sphere containing an inclusion. As the area of the interfaces increases, the backscatter intensity increases, resulting in a decrease in the asymmetry parameter. Specular reflections also occur because of the constructive interference of rays reflecting off the different lattice sites.

14. SUBJECT TERMS
Scatter, FDTD

15. NUMBER OF PAGES
22

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	SAR

NSN 7540-01-280-5500

Standard Form 298 (Rev. 2-89)
Prescribed by ANSI Std. Z39-18
298-102