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Optical characteristics of a cluster of closely-packed dielectric spheres

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Abstract

We employ the pseudospectral time-domain technique to simulate light scattering by a cluster of mono-disperse dielectric spheres. The total scattering cross-section (TSCS) spectrum of the cluster is obtained and analyzed. Research findings show that the TSCS spectrum exhibits characteristics of the cluster geometry as a whole, as well as characteristics of the constituent dielectric spheres. Furthermore, an optical signature indicative of the constituent sphere size is identified, suggesting the possibility to obtain microscopic geometrical information of a closely-packed geometry from multiply scattered light.

Keywords: Multiple scattering; Total scattering cross-section; PSTD; Maxwell's equations

1. Introduction

Light scattering by random media is in general a very difficult problem to analyze, simply due to the enormous number of variables involved. In modern science, astrophysicists have pioneered the research of this problem; their goal was to understand how radiation created in the center of stars is affected when it propagates through interstellar clouds. Chandrasekhar [1] has pioneered in the development of radiative transfer theory, which is the foundation of most heuristic research methods commonly employed nowadays. In the 1950s, many studies have been done concerning the size determination of polystyrene latexes by light scattering; a clear and detailed summary of the theories and various relevant topics was presented by Ishimaru [2]. In acoustics, Twersky [3] has pioneered in the theoretical development for acoustic wave scattering in random media. Mackowski [4] and Mishchenko [4] contributed to the development of the T-matrix method and theories for multiple scattering in clouds. Since then, many researchers in different disciplines have worked on essentially the same problem—light scattering in random media.

Obtaining structural information from scattered light can provide essential information for various applications. In biomedical research, there has been increasing interest in optical diagnostic techniques [5,6], mainly due to the non-invasive characteristics of optical wavelengths. In biomedical applications, sizing of biological structure plays a critical role in determining sub-cellular structures and in vivo detection of morphological changes [7]. In other research areas, optical techniques have also been commonly applied for particle sizing in pharmaceutics and environmental monitoring [8–11]. Generally speaking, the common objective of all these applications is to obtain microscopic structural information from macroscopic scattered light.

Current available optical sizing techniques are mostly based upon single scattering of light [12]. These techniques, such as [10,11], commonly omit contributions of multiply scattering light. Experimental measurements are compared with the Mie theory predictions of a single homogeneous sphere; by comparing the scattered light intensity as a function of angle, an estimate of the scatterer size can be obtained. Such research approaches are most suitable for sparse distributions of scatterers. However, for optically thick structures, light mostly undergoes multiple scattering

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and the assumption of "minimal multiply scattered light" becomes invalid. As a consequence, data interpretation becomes exceedingly difficult for systems with non-negligible contributions from multiple scattering [13]. It certainly would be very helpful if a rigorous research method is available to analyze the characteristics of multiply scattered light.

2. Method

Conventional research methods for the problem of light scattering by macroscopic random medium commonly omit the effect of multiply scattered light [10,11]. To accurately determine the optical characteristics of a macroscopic complex geometry, multiply scattered light must be taken into account. In this Letter, we simulate light scattering by a random medium consisting of randomly positioned dielectric spheres by numerically solving Maxwell's equations. The objective is to investigate the microscopic origin of macroscopic scattered light without heuristic approximations.

The T-matrix method, also known as the transition matrix method, is well known in the quantum theory of scattering which is capable of accurately solving the problem of light scattering by randomly positioned dielectric spheres. For light scattering problems, the T-matrix method first appeared in the pioneer works of P.C. Waterman [15] and later much progress has been made by Mishchenko [4,16,17] and Mackowski [18]. In 2004, Xu [19] reported a generalization of the multi-particle Mie (GMM) expansion, which can predict light scattering characteristics of an ensemble of discrete, nonintersecting particles. Yet, these methods are not applicable to random media in general.

Our long-term goal is to investigate the optical characteristics of macroscopic random media in general, including biological tissues. Therefore, a research method capable of handling random media with continuous variations of the refractive index is required. In this Letter, we employ the pseudospectral time-domain (PSTD) technique pioneered by Liu [14]. The PSTD technique is a variant of the finite-difference time-domain (FDTD) technique. Similar to FDTD, the PSTD technique is also a grid-based technique capable of simulating light scattering by arbitrary geometry. The PSTD technique is advantageous for simulating large-scale electromagnetic problems as it requires less computer memory than the FDTD technique—the PSTD technique reduces the computer memory storage by approximately 8^D:1 (D is the dimension of the problem) relative to the conventional FDTD while achieving comparable accuracy [14]. Thus, the PSTD technique enables a rigorous simulation of a macroscopic light scattering problem, including random media with continuous variations of the refractive index.

For the PSTD simulations, solutions of Maxwell's equations are obtained numerically. The temporal derivatives are calculated using a 2nd-order finite-difference scheme as in the FDTD simulations [20]. The Fourier transforms are employed to calculate the spatial derivatives of the electric and magnetic fields. By transforming the electric and magnetic field components into the frequency domain, the derivatives of the field components can be obtained based on the differentiation theorem of Fourier transform

$$\left\{\left.\frac{\partial \mathbf{E}}{\partial x}\right|_i\right\} = -\mathbf{F}^{-1}(i\tilde{k}_x \mathbf{F}\{\mathbf{E}_i\}) \tag{1}$$

$\{\boldsymbol{E}_i\}$ denotes values of the electric or magnetic field, and $\{(\partial \boldsymbol{E}/\partial x)_i\}$ denotes the spatial derivative of $\boldsymbol{E}$ in the x-direction. The forward and inverse Fourier transforms are represented by $\boldsymbol{F}$ and $\boldsymbol{F}^{-1}$, respectively; $\tilde{k}_x$ is the Fourier transform variable representing the x-component of the numerical wavevector. According to the Nyquist sampling theorem, the spatial derivatives calculated in (1) is of spectral accuracy, enabling the PSTD technique, with a coarse grid of two spatial samples per wavelength, to achieve similar accuracy as the FDTD technique with 20 spatial samples per wavelength.

Lastly, a standard anisotropic perfectly matched layer absorbing boundary condition [21] is implemented to absorb outgoing waves, simulating an open-region light scattering experiment. An impulsive plane wave illuminates the cluster, allowing scattered light of various wavelengths at all angles to be obtained by employing a near-to-far field transformation [20].

The differential scattering cross-section (DSCS) and total scattering cross-section (TSCS) spectra can be obtained from the PSTD simulations according to

$$\mathrm{DSCS} = \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \frac{\text{Scattered flux/Unit of solid angle}}{\text{Incident flux/Unit of surface}} \tag{2}$$

$$\mathrm{TSCS} = \int \mathrm{d}\Omega \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \frac{4\pi}{k}\mathrm{Im} f(0) \tag{3}$$

where $f(0)$ is the scattering amplitude in the forward scattering direction. Eq. (3) is known as the optical theorem [22], which is based upon conservation of energy. With a spatial grid resolution of 0.33 μm, the TSCS spectra of the cluster from 0.5 THz to 300 THz ($\lambda_0 = 600$–1 μm) is calculated, with a spectral resolution of 0.5 THz. For a $(60\ \mu\mathrm{m})^3$ cluster, each PSTD simulation typically takes ~12 h with a parallel computer cluster of twenty 2.4-GHz Pentium-4 Xeon processors.

3. Results

A validation of the PSTD simulation of light scattering is shown in Fig. 1. Light scattering by a dielectric sphere with refractive index $n = 1.2$ sphere is simulated. DSCS as a function of angle is obtained and compared with the Mie expansion [23]. It is readily seen that the PSTD-computed scattering characteristics show good agreement with the analytical expansion of Maxwell's equations.

Next, we simulate light scattering by a cluster of dielectric spheres and calculate its spectral scattering characteristics. As shown in Fig. 2, the TSCS spectrum of a cluster of

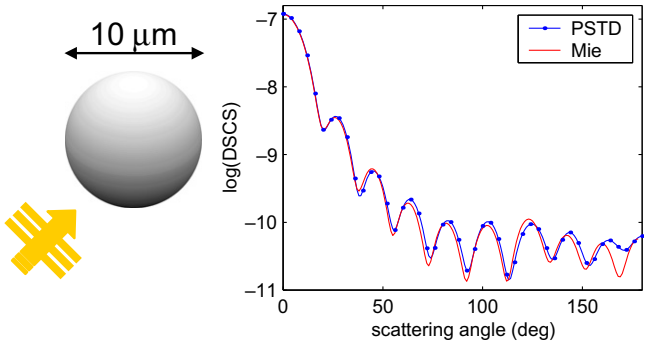

Fig. 1. Validation of the 3-D PSTD light scattering simulations. Light scattering by a homogeneous dielectric sphere with a refractive index $n = 1.2$ is simulated using the PSTD technique with a grid resolution of $dx = 0.33$ μm. The DSCS as a function of angle is obtained and compared with the Mie expansion.

dielectric spheres is determined by employing the PSTD simulations. Notice that with more spheres closely-packed within the cluster, the TSCS spectrum evolves: a peak of the TSCS spectrum gradually emerges around 40–50 THz as the number of spheres within the cluster increases. This TSCS peak corresponds to the Mie resonance of the cluster geometry as a whole. In the long-wavelength limit, the incident light is insensitive to geometrical structures much smaller than the wavelength; in other words, the incident light cannot "see" geometrical structures on a length scale shorter than the wavelength. As a result, the TSCS spectrum exhibits optical characteristics partially due to the cluster as a whole, and partially due to the individual constituent dielectric spheres.

In order to extract geometric information indicative of the cluster geometry, we employ a cross-correlation analysis. By calculating the cross-correlation coefficient of the cluster TSCS spectrum and the TSCS spectrum of a single d-μm-diameter dielectric sphere, the TSCS spectral relevance can be quantified. If the cluster TSCS spectrum exhibits characteristics of a d_p-diameter sphere, the cross-correlation coefficient should reach its maximum value at d_p. Hence, the cross-correlation analysis enables matching the relative amplitude and frequency variations, and can quantify the relevance and determine the best match for the cluster TSCS.

We further employ the cross-correlation analysis to analyze the light scattering characteristics of mono-disperse, d-μm-diameter dielectric spheres. The specific aim is to test whether the cross-correlation analysis is applicable to other diameter spheres in general. As shown in Fig. 3a–d, the estimated diameter d_p yielded by the cross-correlation analysis varies monotonically relative to the actual diameter of the constituent spheres. Specifically, d_p is approximately equal to the actual diameter of the constituent spheres, as shown in Fig. 3e.

Lastly, we investigate the effect of optical thickness on the cross-correlation analysis. The following definition for optical thickness is used: optical thickness ≡ (geometrical thickness)/(scattering mean free path). The TSCS spectra corresponding to clusters of different number of spheres (N) are obtained from the PSTD simulations, as shown in Fig. 4. For small values of N, the TSCS spectrum is similar to the TSCS of a single dielectric sphere, whereas for larger N, the multiple scattering effect becomes more

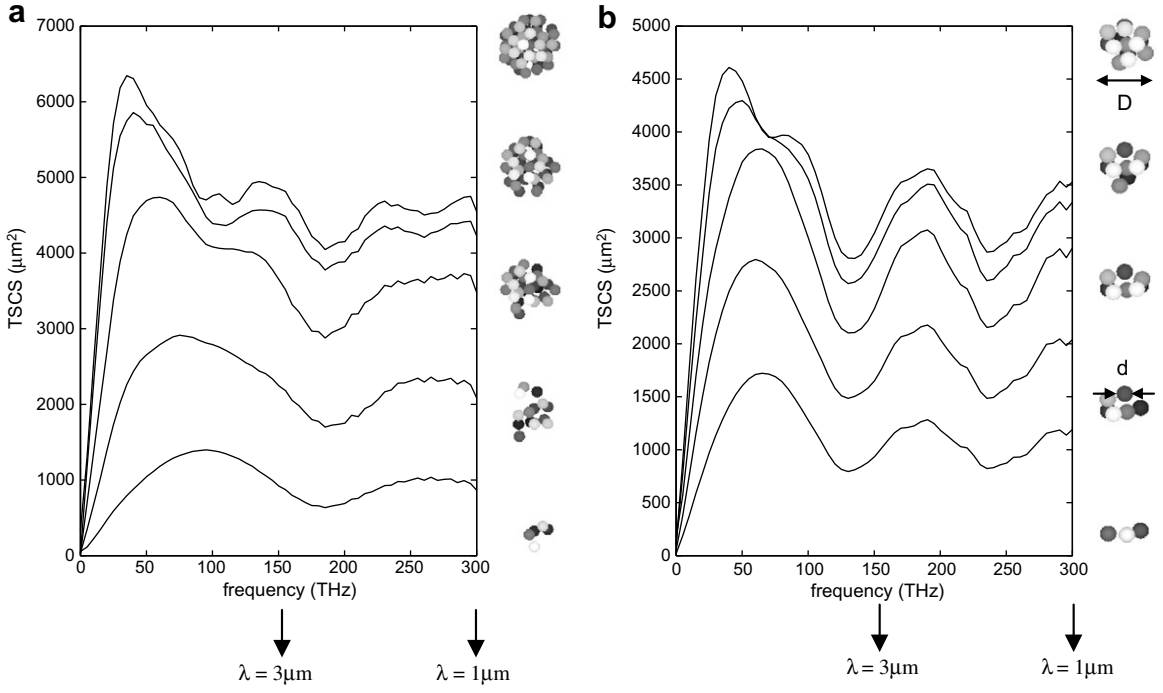

Fig. 2. Spectral light scattering characteristics of a cluster of dielectric spheres is determined by means of a PSTD simulation. With an overall diameter $D = 50$ μm, each cluster consisting of N randomly positioned, non-contacting, $n = 1.2$ dielectric spheres of individual diameter d μm. The TSCS spectra of the cluster geometry are obtained. Five cases are shown (bottom to top): (a) $d = 10$ μm, $N = 5$, 15, 30, 50, and 56; (b) $d = 14$ μm, $N = 3$, 6, 8, 10, and 14.

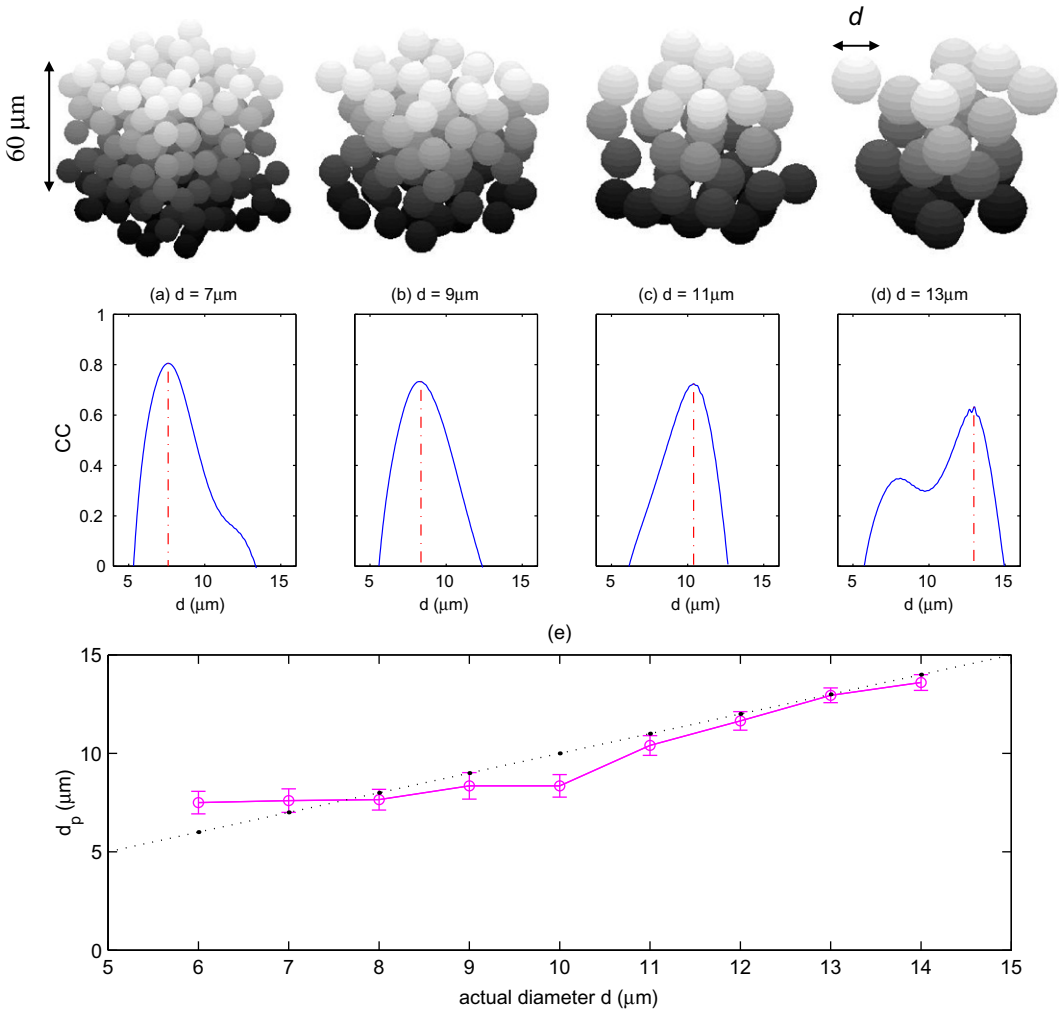

Fig. 3. Cross-correlation analysis of the cluster TSCS spectra of a cluster of d-μm-diameter dielectric spheres. Each $(60\ \mu m)^3$ cluster consists of N randomly positioned, closely-packed, $n = 1.2$ dielectric spheres: (a) $d = 7$ μm, $N = 192$, (b) $d = 9$ μm, $N = 86$, (c) $d = 11$ μm, $N = 42$, (d) $d = 13$ μm, $N = 24$. The correlation coefficient (CC) peaks at d_p; d_p is plotted in (e) and compared with the actual diameter d. (Each error bar represents the CC peak width at 80% of the peak amplitude.)

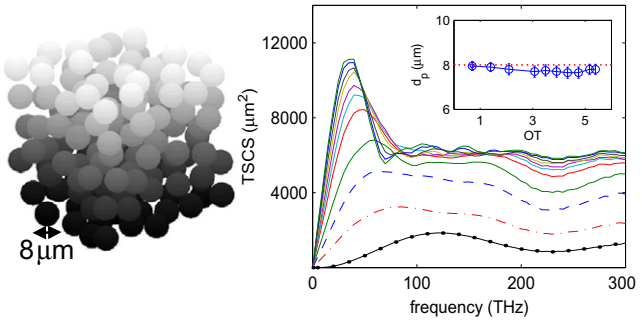

Fig. 4. PSTD-computed TSCS spectra of a $(60\ \mu m)^3$-cluster consisting of N of randomly positioned, 8 μm-diameter, $n = 1.2$ dielectric spheres. From bottom to top: $N = 1$, 25, 50, 75, 110, 125, 140, 155, 170, 185, and 193. (The TSCS spectrum of a single 8 μm-diameter sphere ($N = 1$) is magnified by 10 times to facilitate comparison.) d_p from the cross-correlation analysis is plotted versus optical thickness (OT) of the cluster as shown in the inset.

pronounced, and the TSCS characteristics due to the cluster as a whole emerge. Yet, even though the TSCS characteristics due to the constituent spheres are obscured by the characteristics due to the overall geometry, the estimated sphere diameter d_p as determined by the cross-correlation analysis shows excellent agreement with the actual diameter d.

4. Discussion

The simulations reported in this Letter are idealized virtual experiments in a practically noise-less environment. In each simulation, a linearly polarized light is employed as the light source. Since the geometry consists of homogeneous and isotropic dielectric spheres, it is anticipated that circularly polarized light illumination will give results similar to linearly polarized illumination. As for the refractive index, a typical refractive index of common dielectric materials ranges from 1 (vacuum) to 2 (crystal). We have chosen a representative refractive index $n = 1.2$ for the purpose of this research; nevertheless, simulations have been done for other refractive indices and showed similar results.

We report discovery of a spectral signature of the TSCS spectrum that is indicative of the specific microscopic geometry of closely-packed dielectric spheres. The underlying mechanism of the TSCS spectral signature is not fully understood. We suspect this may be analogous to the phenomenon of resonance—the cluster of dielectric spheres reacts differently to specific incident wavelengths that match with the characteristic lengths of the cluster (e.g., diameter of the constituent spheres). However, exactly how the cluster would react to specific wavelengths is yet to be determined. Could information regarding the microscopic geometry of the random medium be extracted from the TSCS spectrum? Specific results reported in this Letter may provide insight to answering these questions.

5. Conclusion

In this Letter we show that the optical characteristics are contributed both from the cluster geometry as a whole as well as from the individual constituent spheres: in the long-wavelength regime, the TSCS spectrum is dominated by the overall geometry; whereas in the short-wavelength regime, the TSCS spectrum exhibits characteristics due to the individual constituent spheres. Specific results show a correlation relationship between the macroscopic scattered light and its microscopic origin, it is clear that the multiply scattered light contains information indicative of the size of constituent spheres of the cluster. This information can be extracted by means of a cross-correlation analysis. More generally, our research methodology is applicable to arbitrary geometry, including random medium of continuous variations of refractive index.

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