Journal of Quantitative Spectroscopy & Radiative Transfer 196 (2017) 94–102

Contents lists available at ScienceDirect

Journal of Quantitative Spectroscopy & Radiative Transfer

journal homepage: www.elsevier.com/locate/jqsrt

Dependent scattering and absorption by densely packed discrete spherical particles: Effects of complex refractive index

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ARTICLE INFO

Article history:
Received 21 February 2017
Received in revised form
22 March 2017
Accepted 29 March 2017
Available online 5 April 2017

Keywords:
Radiative transfer
Maxwell equations
Dependent scattering
T-matrix method
Mueller matrix

ABSTRACT

Due to the dependent scattering and absorption effects, the radiative transfer equation (RTE) may not be suitable for dealing with radiative transfer in dense discrete random media. This paper continues previous research on multiple and dependent scattering in densely packed discrete particle systems, and puts emphasis on the effects of particle complex refractive index. The Mueller matrix elements of the scattering system with different complex refractive indexes are obtained by both electromagnetic method and radiative transfer method. The Maxwell equations are directly solved based on the superposition T-matrix method, while the RTE is solved by the Monte Carlo method combined with the hard sphere model in the Percus-Yevick approximation (HSPYA) to consider the dependent scattering effects. The results show that for densely packed discrete random media composed of medium size parameter particles (equals 6.964 in this study), the demarcation line between independent and dependent scattering has remarkable connections with the particle complex refractive index. With the particle volume fraction increase to a certain value, densely packed discrete particles with higher refractive index contrasts between the particles and host medium and higher particle absorption indexes are more likely to show stronger dependent characteristics. Due to the failure of the extended Rayleigh-Debye scattering condition, the HSPYA has weak effect on the dependent scattering correction at large phase shift parameters.

1. Introduction

The problem of light scattering and absorption by densely packed discrete random media is important in many science and engineering fields, such as remote sensing data analysis in astrophysics and atmosphere science, light interaction with tissues in biomedical diagnostics, and radiative heat transfer in chemical reactors, coal combustors and many other industrial systems [1–3]. In the theoretical analysis of radiative transfer process in a dense discrete random medium, one important characteristic is that the dependent scattering and absorption effects cannot be neglected in many cases, which make it become a much more difficult task than in a dilute medium. The dependent effects are mainly caused by the near-field interparticle effect which modifies the amount of radiation absorbed and scattered by each particle and the far-field interference effect which leads to a coherent addition of the scattered radiation by each particle in the far field [4,5]. For a dilute medium, each particle is assumed to be located in the far-field zones of all the other particles and acts independently in the scattering and absorption of radiation. For light transport in a dilute medium, the radiative transfer theory (RTT) has been a most effective and widely used approach [6]. However, the radiative transfer equation (RTE) will fail for wavelength sized particles with concentrations increasing to a certain value due to the dependent scattering and absorption effects [4,7]. Despite the obvious shortcomings of the RTE, its applications appears to be unavoidable in many cases. The main cause of this situation is the limited application of direct computer solvers of the Maxwell equations to realistic media composed of extremely large numbers of particles [1,6].

At present, the researches on the radiative transfer in dense discrete random media mainly focus on two aspects: to determine the demarcation line between independent and dependent scattering and to improve the validity of the RTE by considering the dependent scattering and absorption effects [5,8–14]. Drolen et al. [8,9] constructed a widely used regime map demarcating independent and dependent scattering over a wide range of size parameters and volume fractions based on theoretical analysis and experimental research. The results showed that the criteria of

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http://dx.doi.org/10.1016/j.jqsrt.2017.03.039
0022-4073/

dependent scattering mainly depend on the ratio of the interparticle clearance c and wavelength λ. Hespel et al. [10] studied the extinction coefficient of latex suspensions composed of densely packed dielectric spheres through experiments. Their results indicated that the demarcation line (c/λ=0.5) between independent and dependent scattering is unrealistic except for small size parameters. Kumar and Tien [5] presented a complete model to evaluate the dependent scattering and absorption characteristics of dense particulate systems. However, the research only focuses on the small particles in the Rayleigh scattering regime. Tishkovets and Petrova [11] considered the densely packed discrete random media as semi-infinite layer composed of randomly oriented clusters. The RTE is then used to obtain the reflection characteristics of the medium by assuming that the clusters are in the far zones of other clusters. Mishchenko et al. [12] assessed the application scope of the vector radiative transfer equation (VRTE) by comparing with Stokes reflection matrix of latex suspensions obtained by experiments. The paper demonstrated that a simple modification of the phase matrix based on the static structure factor in the Percus-Yevick approximation yields an improved fit to the experimental results.

Although many experimental researches on radiative transfer in dense discrete random media have been carried out, due to the limits of material type of standard particles, only a few types of materials are used, such as polystyrene and silicon dioxide [7,9,10,12,13,15–18]. Moreover, few researchers had investigated the effect of complex refractive index of particles on the light scattering process in dense discrete random media, especially when the polarization effects and dependent effects correction are taken into consideration.

In this paper, the effects of particle complex refractive index on dependent scattering and absorption by densely packed discrete spherical particles are investigated as an extension of our previous researches which examined the accuracy of RTE by comparing with the exact electromagnetic theory and focused on the influences of particle volume fractions and particle size parameters [19]. The dependent scattering effects are considered into the RTE based on the hard sphere model in the Percus-Yevick approximation (HSPYA). The Mueller matrix elements of the system are used for assessing the validity of the RTE and the performance of dependent scattering correction.

2. Modeling methodology

We consider light scattering and absorption in an imaginary spherical volume filled with randomly distributed, densely packed spherical particles. The scattering volume is illuminated by a parallel quasi-monochromatic beam of light and the observation point is located in the far-field zone of the entire volume. In this study, the Mueller matrix of the scattering system is obtained by both the electromagnetic method and radiative transfer method. The Mueller matrix **M** links the Stokes vectors of scattered light $\mathbf{I_{sca}(\Omega)}=(I_{sca}, Q_{sca}, U_{sca}, V_{sca})^T$ in the scattering direction $\mathbf{\Omega}$ and incident light $\mathbf{I_{inc}(\Omega')}=(I_{inc}, Q_{inc}, U_{inc}, V_{inc})^T$ in the incident direction $\mathbf{\Omega'}$, and can be written as [6,20]:

$$\begin{bmatrix} I_{sca} \\ Q_{sca} \\ U_{sca} \\ V_{sca} \end{bmatrix} \propto \begin{bmatrix} M_{11}(\theta) & M_{21}(\theta) & 0 & 0 \\ M_{21}(\theta) & M_{22}(\theta) & 0 & 0 \\ 0 & 0 & M_{33}(\theta) & M_{34}(\theta) \\ 0 & 0 & -M_{34}(\theta) & M_{44}(\theta) \end{bmatrix} \begin{bmatrix} I_{inc} \\ Q_{inc} \\ U_{inc} \\ V_{inc} \end{bmatrix}, \quad (1)$$

where $\theta\in[0°, 180°]$ is the angle between the incidence and scattering directions, as seen in Fig. 1. In Eq. (1), the zeros denote the Mueller matrix elements which are negligibly small relative to the

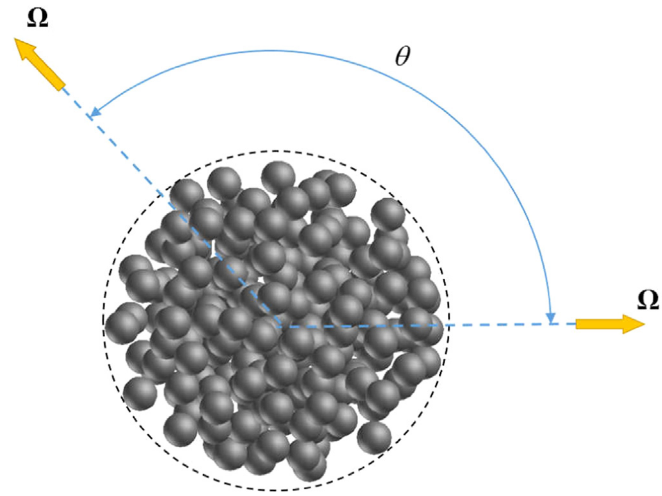

Fig. 1. Schematic diagram of the electromagnetic scattering model.

other elements. The (1,1) elements, $M_{11}(\theta)$, which is called phase function is normalized according to

$$\frac{1}{2}\int_0^{\pi} M_{11}\sin\theta d\theta = 1. \quad (2)$$

In addition, all other Mueller matrix elements are normalized to the element M_{11} at the same angle to get results within a range from −1 to 1.

The Maxwell equations are directly solved by using the highly efficient and numerically exact superposition T-matrix method (STMM) [21,22]. To generate ensembles of randomly distributed, densely packed spheres in an imaginary spherical volume, the Metropolis shuffling algorithm is used [23]. Note that the spheres in the simulation volume are not allowed to be overlapped and cross the volume's outer boundary. By taking the sphere positions and sphere size parameters as input, the Mueller matrix can be directly calculated by using the STMM program [22].

The RTE is numerically solved by using the Monte Carlo (MC) method [24–26]. Before the MC simulation, the radiative properties of the spheres should be calculated. In the framework of independent scattering theory, the radiative properties which include scattering coefficient μ_{sca}, absorption coefficient μ_{abs}, and single scattering Mueller matrix **P** are calculated using the Lorenz-Mie theory [27,28]. For monodispersed spheres, the scattering coefficient μ_{sca} and absorption coefficient μ_{abs} are calculated as

$$\mu_{sca} = 0.75Q_{sca}\frac{f_v}{a}, \quad (3)$$

$$\mu_{abs} = 0.75Q_{abs}\frac{f_v}{a}, \quad (4)$$

where a and f_v are the sphere radius and sphere volume fraction, Q_{sca} and Q_{abs} are the scattering efficiency and absorption efficiency determined from calculations [27]. For densely packed spheres with high volume fractions, the far-field interference interaction between particles occurs and may result in an essential alteration of the scattering properties. Based on the research of Cartigny et al. [4] and Tuchin et al. [3], the dependent scattering efficiency $Q_{sca,d}$, dependent scattering coefficient $\mu_{sca,d}$, and dependent single-scattering Mueller matrix elements $P_{ij,d}$ can be calculated as

$$\frac{Q_{sca,d}}{Q_{sca}} = \frac{1}{2}\int_0^{\pi} S(f_v, \theta)P_{11}(\theta)\sin d\theta d\theta, \quad (5)$$