Direct Simulation of Scattering and Absorption by Particle Deposits

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1 Introduction

The present paper is concerned with the prediction of the radiative absorption and scattering behavior of a layer of densely packed, wavelength–sized particles. This problem has relevance to a number of engineering and scientific applications, e.g., estimation of the effect of particle deposits on heat exchanger surfaces, modeling the reflection properties of pigment coatings and dust layers, and prediction of Anderson localization in discretely inhomogeneous media.

A well–developed understanding exists for calculation of the single (or isolated) scattering and absorption properties of particles – encompassing simple shapes (such as Lorenz–Mie theory for spheres) to more sophisticated methods for nonspherical and inhomogeneous particles [1]. In the situations of interest here, e.g., particle deposits, paint pigments, composite materials, etc., the particle concentrations can become sufficiently high so that prediction of the particle optical properties via single–scattering formulations becomes suspect. Specifically, under such conditions the electric field incident on a particle can have significant contributions due to scattering from neighboring particles (so–called near field interactions) and the far field scattering can be modified by the correlated positions existing among the close–packed particles (far field interference). These two effects are typically referred to together as dependent scattering, and generally become significant for particle volume fractions $f > 0.01$ and/or particle clearance/wavelength ratios less than 0.5 [2]; such conditions typically involve packed deposits of particles having size parameters $x = 2\pi a/\lambda$ (where a is a characteristic radius of the particle and λ is the radiation wavelength) on the order of unity or less.

Initial investigations on dependent scattering primarily dealt with the propagation of a coherent wave through a particulate medium, with the objective of identifying an effective propagation constant (or, equivalently, complex refractive index) of the medium which describes the attenuation of the coherent wave via absorption and scattering by the particles [3, 4]. In principle, it is possible to exactly calculate, via analytical superposition methods, the absorption and scattering properties of neighboring particles provided the single–scattering properties are known. A review of the superposition method, as applied to spherical particles, is given in Ref. [5]. Until now, the application of the superposition method has primarily been to determine the optical properties of aggregated particles containing a finite number of spheres. A few investigations have been conducted which compared direct simulations of wave propagation in large ensembles of spheres – as exactly calculated with the superposition method – to effective medium theories [6, 7]. Effective medium theories have also been coupled to the superposition method, with the objective of developing efficient methods for computation of scattering properties of nonspherical particles [8, 9].

The objective of this paper is to demonstrate the feasibility of using exact methods to directly simulate the absorption and scattering properties of a plane layer of densely–packed spheres that are exposed to an incident source of radiation. Such methods could provide benchmark calculations for gauging the accuracy of effective medium/radiative transfer equation (RTE) models as applied to thin deposits or coatings of particles. Since the exact methods also provide a detailed description of the electric field distribution both within and external to the particles, calculations of this sort would also be useful in the examination of localization phenomena in random media. The motivation for this approach stems from the fact that the computational algorithms for wave interactions among spheres have progressed to the point that direct calculations involving large–scale ensembles – i.e., sphere systems that are adequately large to represent a radiative continuum – have become tractable on typical desktop PCs [5].

The particle layer in this investigation will be represented by a large, yet obviously finite, number of spheres that are arranged in set positions. The thickness of the layer will be a fixed parameter in the simulations, yet the lateral extent should be sufficiently large to represent an infinite expanse of particles. Meeting this condition is difficult – if not impossible – when the 'target' of spheres is exposed to a transverse plane wave. In this case the lateral extent of the layer will always have an effect on the far–field scattering pattern, due to diffraction at the edges. This problem is bypassed by using a focussed beam of radiation as the exciting (or probing) source, in which the width of the beam is smaller than the lateral size of the layer.

2 Formulation

The system under examination consists of an ensemble of N_S spheres, each characterized by a size parameter $x_i = \mathrm{k}a_i$, a complex refractive index $m_i = n_i + ik_i$, and a position (X_i, Y_i, Z_i) for $i = 1, 2, \ldots N_S$. The incident field consists of a focussed beam which propagates in a direction $\theta = \beta_0$ and $\phi = \alpha_0$ relative to the target coordinate frame and is focussed at a point X_0, Y_0, Z_0. Along the focal plane (which contains the focal point and is perpendicular to the propagation direction) the beam is approximated as a linearly polarized transverse wave, i.e.,

$$\mathbf{E}_{inc}(X', Y', 0) \approx \hat{\mathbf{x}} \exp\left(-\frac{X'^2 + Y'^2}{\omega_0^2}\right) \tag{1}$$

in which ω_0 is the beam width parameter and the primed coordinates denote the rotated coordinate system that is centered on the focal point and with a z' axis pointing in the propagation direction.

The solution method used to obtain the scattered field is a direct extension of Lorenz/Mie theory. The total field external to the spheres is represented as a sum of fields scattered from the individual spheres in the ensemble plus the incident field;

$$\mathbf{E}_{ext} = \mathbf{E}_{inc} + \sum_{i=1}^{N_S} \mathbf{E}_{sca,i} \tag{2}$$

The incident field is represented as a regular vector spherical harmonic (VSH) expansion centered about an arbitrary origin, whereas each of the scattered fields is represented by an outgoing VSH expansion centered about the origin of the sphere. Application of the continuity equations at the surface of each sphere, and utilization of the addition theorem for VSH, results in a system of equations for the expansion coefficients for the individual scattered fields;

$$\frac{1}{\overline{a}_{np}^i}\, a_{mnp}^i - \sum_{\substack{j=1 \\ j\neq i}}^{N_S} \sum_{n'=1}^{L_i} \sum_{m'=-n'}^{n'} \sum_{p'=1}^{2} H_{mnp\,m'n'p'}^{i-j}\, a_{m'n'p'}^j = g_{mnp}^i \tag{3}$$

In the above, a_{mnp}^i and g_{mnp}^i denote the expansion coefficients, of order n, degree m, and mode p $(= 1, 2$ for TM/TE) for the scattered and incident fields centered about sphere i, H^{i-j} is a translation matrix which transforms an outgoing VSH centered about origin j into an expansion of regular VSH about i, and $\overline{a}_{np}^i$ denote the Lorenz/Mie coefficients for sphere i, which are a function of the sphere size parameter x_i and refractive index m_i.

The present application is concerned primarily with the propagation of a collimated beam into a particulate medium. Accordingly, conditions are sought which minimize the spreading of the beam waist as a function of Z'. Such conditions will correspond to relatively large $\mathrm{k}\,\omega_0$, which is equivalent to a large diffraction length/spot size ratio $(2\pi\omega_0^2/\lambda)/\omega_0$. Fortunately, such conditions also allow a relatively simple formula for the focal–point centered VSH expansion coefficients for the incident beam via the localized approximation. The expansion coefficient for the beam centered about a sphere origin i, i.e., g_{mnp}^i, can then be obtained by application of the VSH addition theorem [10, 11].

The complete scattering and absorption properties of the system can be obtained from the solution to Eq. (3). Such properties include the absorption cross sections of the individual spheres and the bulk absorptive and reflective properties of the slab as a whole.

3 Results and Discussion

The target used in this work consisted of a cylindrical slab of spheres that were packed into a tetrahedral lattice. The axis of the cylinder is taken to be the Z direction, and the radius (which is proportional to the

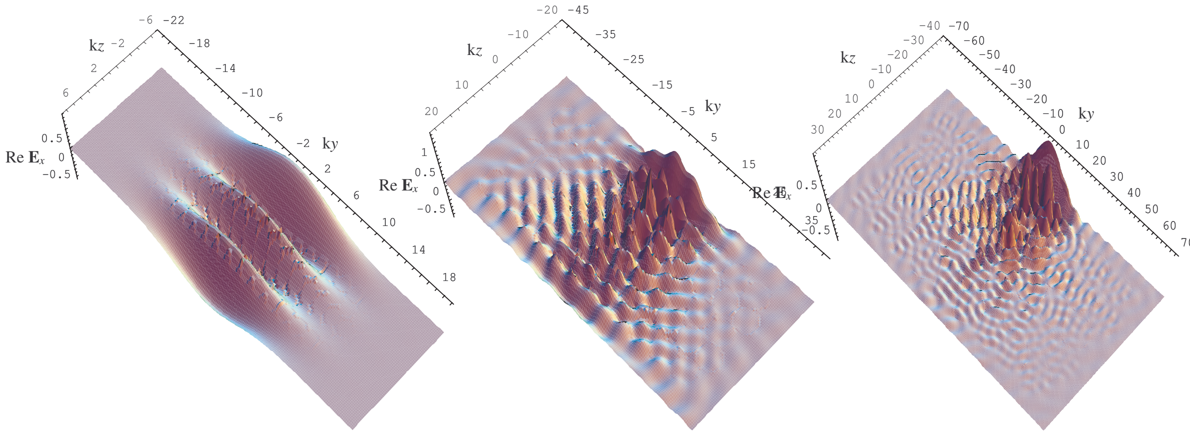

Figure 1: Re $(\hat{\mathbf{x}} \cdot \mathbf{E})$ vs. position on the $Y' - Z'$ plane. Sphere size parameter $x_S = 1$ (l), 4 (c), 8 (r), refractive index $m = 1.6 + 0.02i$.

number of spheres in the X and Y directions) is chosen to be several times larger than the incident beam width ω_0. The incident beam was characterized by a dimensionless beam width parameter of $\mathrm{k}\,\omega_0 = 10$.

Shown in Fig. (1) are surface plots of the X–component of electric field in the $Y - Z$ plane, calculated for sphere size parameters of $x_S = 1$, 4, and 8. The sphere refractive index is $m_S = 1.6 + 0.02i$ for all cases. The sphere matrix consists of 5 sphere layers in the Z direction, and the incident beam is focussed in the center of the matrix and was incident parallel to the z axis (representing normal incidence on the slab). The number of spheres in the slab depended on the size parameter, with a smaller x_S requiring more spheres in order to extend the slab radius past the width of the beam. For $x_S = 1$ and 8 the model required 1760 and 275 spheres. The latter case used a truncation order of $L = 11$ for the sphere scattered field expansions, which corresponded to 44,000 complex–valued equations for the set of scattering coefficients in Eq. (3). Electric field components were calculated using the superposition of Eq. (2) if the point was external to the spheres, or using the Lorenz/Mie relations to relate the internal to external fields for points interior to the spheres.

One point to make regarding Fig. (1) is that it demonstrates the veracity of the formulation and computations. The particular plane chosen for the surface plots splits the spheres intersected by the plane in half, and accordingly the sphere surfaces will be tangential to the x direction along the plane. Since the tangential components of electric field are continuous at the sphere surface, the plots should demonstrate a continuity in electric field from the exterior to interior regions. This behavior is completely consistent with the calculation results. Relatively close inspection of Fig. (1) is needed to discern the sphere positions by virtue of the small jumps in electric field that occur at the sphere edges, which are due to truncation errors in the series solution (most noticeably for the $x_S = 1$ case).

The results in Fig. (1) show that the slab of spheres with $x_S = 1$ behaves as a homogeneous medium, in that the profile of the incident beam is largely unperturbed as it propagates through the slab. This behavior is somewhat surprising: an effectively homogeneous medium would be expected for $x_S \ll 1$, for which the simulation would become equivalent to a discrete dipole model for a homogeneous slab, yet the $x_S = 1$ spheres do not behave as dipoles. Indeed, three harmonic orders (N_O) were needed to represent the scattered fields from the spheres, as opposed to a single TM order for the dipole. A relevant condition behind the apparent homogenous behavior has to do with the fact that the sphere radius, for this case, is significantly smaller than the width of the incident beam. Because of this, a relatively large population of spheres are excited by the beam. The scattered field produced by the spheres – and the resulting interference of fields – will therefore be averaged over the large group of scattering sources, resulting in a net field which is not strongly dependent on position in the slab. This behavior is in keeping with the Quasi–Crystalline Approximation in effective medium models [4].

Conversely, the results for $x_s = 4$ and 8 show that the field within the slab for larger size parameters can become highly dependent on position. The peaks in the field amplitudes are associated with the focussing of internal fields within the individual spheres, and this effect becomes more pronounced as the size parameter increases. For both $x_S = 4$ and 8, the effects of multiple scattering among the spheres leads to a broadening – or diffusion – of the field with increasing depth into the medium. For the smaller size parameter the field distribution remains symmetrical in the $y - z$ plane – which would be expected due to the symmetrical conditions imposed on the problem – yet for $x_S = 8$ the field distribution appears to take on a chaotic structure.

The distribution of absorption in the medium – which allows for determination of the bulk absorption coefficient – as well as the reflectivity of the slab and the far–field scattering behavior will be presented at the meeting.

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