MULTIPLE SCATTERING BY DENSELY PACKED DISCRETE RANDOM MEDIA: EXACT RESULTS.

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Introduction: Multiple scattering of electromagnettic waves by macroscopic media composed of randomly positioned particles is a subject of great importance to many science and engineering disciplines. Until quite recently, the only practical means of multiple-scattering computations for turbid and other particulate media have been various approximate approaches such as the effective field approximation, the quasi-crystalline approximation, the diffusion approximation, the radiative transfer theory (RTT), the phenomenological and microphysical theories of coherent backscattering (CB), and the phenomenological theory of strong localization. However, the ever increasing power of scientific workstations and the availability of efficient numerical techniques have recently led to the emergence of an accurate quantitative approach to this complex problem based on direct computer solutions of the Maxwell equations. For practical reasons, this approach cannot be used yet to simulate electromagnetic scattering by random media consisting of extremely large numbers of particles such as clouds, colloids, and powder surfaces. However, it does provide the potential to model rather complex particulate systems and determine all quantitative scattering characteristics some of which may not be straightforward to measure accurately. Therefore, this approach can be used to evaluate the predictions and conditions of applicability of an approximate theory.

The objective of this talk is to use numerically exact solutions of the Maxwell equations in order to simulate the effect of randomness of particle positions and the onset and evolution of multiple-scattering effects with increasing number of particles randomly distributed throughout a finite scattering volume. This allows us to model and analyze the specific scattering regimes that are encountered in such disciplines as dynamic light scattering, RTT, and the theory of CB.

All numerical results described in this talk have been obtained with the highly efficient superposition *T*-matrix method (STMM) [1]. Although the STMM has been used extensively in computations for particle aggregates such as fractal clusters composed of touching soot or mineral monomers, the approach adopted for this study is to model specifically light scattering by a statistically homogeneous volume of particulate medium. Keeping the size of the volume fixed and gradually increasing the number of randomly distributed particles allows us to perform a systematic analysis of emerging and intensifying multiple-scattering effects and thereby illustrate and substantiate the specific assumptions used in the microphysical derivation of the radiative transfer equation (RTE) and in the microphysical theory of CB.

Results and Discussion: We assume that a number N of identical spherical particles are distributed randomly throughout a spherical volume V with a radius R much greater than the particle radius a, as shown in Fig. 1. The size parameter of the particles is fixed at ka = 4, whereas the size parameter of the spherical volume is fixed at kR = 40, where k is the wave number in the surrounding medium. The refractive index of the particles relative that of the host medium is 1.32.

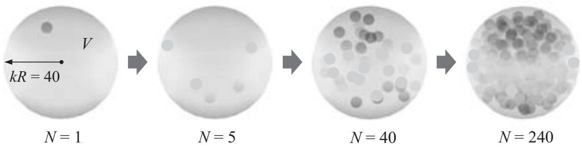

Fig. 1. N particles are distributed randomly throughout a spherical volume V of the host

In order to simulate measurements of light scattering by a rapidly changing object one needs to solve the Maxwell equations repeatedly for a representative set of distinct object configurations. After the set of solutions of the Maxwell equations has been obtained, one has a choice of (i) analyzing the statistical information content of differences in the individual solutions or (ii) applying an averaging procedure and thereby isolating the static component of the scattering pattern. These two approaches are known as *dynamic* and *static* light scattering.

To simulate static light scattering, one needs an efficient way of averaging the computed scattering signal over very many configurations of the N-particle group. A brute-force solution would be to use a random coordinate generator repeatedly to create a large number of different N-particle configurations and then average numerically the corresponding T-matrix results. The more effective approach used here is to create only one random N-particle configuration and then average over all possible orientations of this configuration with respect to the laboratory coordinate system. This procedure yields an infinite continuous set of random realizations of the N-particle group and takes full advantage of the highly efficient orientation averaging procedure afforded by the STMM.

Let us assume that the large spherical volume V is illuminated by a plane electromagnetic wave. The incidence direction coincides with the positive direction of the z-axis of the laboratory reference frame and the meridional plane of the incidence direction coincides with the xz half-plane with $x > 0$. The angular distribution and polarization state of the scattered light in the far-field zone of the entire

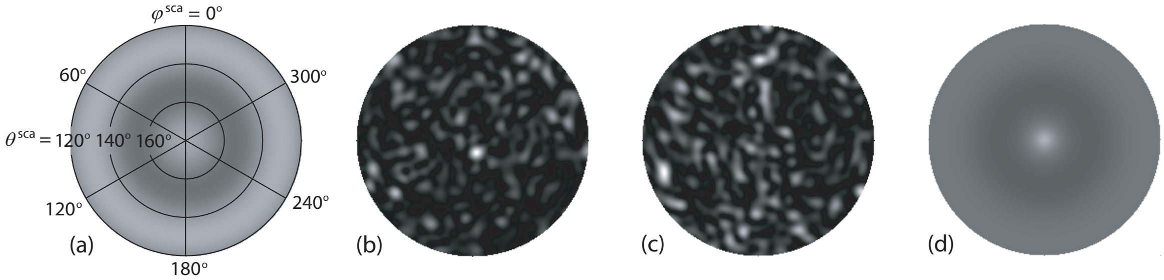

Fig. 2. Angular distribution of scattered intensity in the far-field zone of the spherical volume *V* filled with *N* particles. (a) *N* = 1, fixed orientation. (b) and (c) *N* = 80, fixed orientation. (d) *N* = 80, random orientation. The gray scale is individually adjusted in order to maximally reveal the details of each scattering pattern. Panel (a) also shows the angular coordinates used for all panels.

scattering volume is described by the Stokes phase matrix.

Let us first assume that the incident light is circularly polarized in the counter-clockwise sense when viewed in the direction of propagation, Panels (a) and (b) of Fig. 2 show the far-field angular distributions of the intensity scattered in the backward hemisphere by the large spherical volume filled with *N* = 1 and 80 particles having the same refractive index *m* = 1.32. The individual particle positions were chosen randomly using a random coordinate generator, but otherwise they are fixed. The scattering pattern for *N* = 1 is rather smooth and perfectly azimuthally symmetric, as it should be for a single wavelength-sized spherical particle. However, panel (b) demonstrates a typical speckle pattern.

Of course, the speckle pattern depends not only on the number of particles *N* but also on the specific way they are arranged with respect to the laboratory coordinate system. This is illustrated by panels 2b and 2c computed for two different random 80-particle configurations.

Figures 2b and 2c illustrate the range of variability of the speckle pattern that can be expected upon even minute changes in a random multi-particle configuration. Obviously, neither the speckle pattern nor its variability are reproduced by the classical theories of RT and CB, which indicates that *neither theory describes the instantaneous state of electromagnetic radiation* in a discrete random medium. Instead, both theories fall in the realm of static scattering and describe the result of averaging the relevant optical observables over a significant period of time or, equivalently, over a significant range of random particle positions.

To illustrate this fundamental point, Fig. 2d shows the result of averaging the speckle pattern over the uniform orientation distribution of the 80-particle configuration used to compute Fig. 2b. One can see that with the exception of a notable backscattering peak, the speckle structure is essentially gone. This is not surprising. Indeed, each speckle element is the result of constructive or destructive interference of two wavelets scattered along specific particle sequences. The phase difference between the wavelets changes randomly as the particles move, so that the average result of the interference is zero. However, there are certain pairs of wavelets that interfere constructively irrespective of particle positions and thereby are responsible for the residual scattering pattern. We will demonstrate in this talk that the backscattering intensity peak seen in Fig. 2d as well as the smooth intensity background are in fact caused by special classes of such wavelet pairs [2].

References: [1] Mishchenko M. I., et al. (2002) *Scattering, Absorption, and Emission of Light by Small Particles* (Cambridge University Press, 2002). [2] Mishchenko M. I., (2008): *Rev. Geophys.* **46**, doi:10.1029/2007RG000230.