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Scattering by Rough Particles and Stochastic Aggregates

Abstract

We consider light scattering by three classes of particles: polyhedral particles, stochastic solids and stochastic aggregates. Many naturally occurring particles can be modeled by these classes. Basic light scattering models for these types of particles are reviewed. We present some results for stochastically rough particles and particle aggregates. The calculations have been carried out both by rigorous wave optical methods and by geometric optics approximations. One of the main goals is to study whether these two approaches produce similar results at some intermediate size parameter range. We also propose laboratory measurements for sand particles and algae to test some of our methods.

1 Introduction

A vast amount of theoretical calculations have been made for light scattering by perfect spheres to explain either laboratory measurements or to interpret astronomical data. However, this uncritical use of Mie theory is known to lead to spurious and completely misleading results, although the fits might formally look perfect. It is only quite recently that new and rigorous methods have become available to model light scattering by naturally occurring particles and particle aggregates. To mention just a few, the discrete-dipole approximation (DDA) and the T-matrix methods allow us to calculate light scattering by small particles (size parameter $x \lesssim 20$) almost exactly once the scattering geometry has been correctly modeled. On the other end, geometric

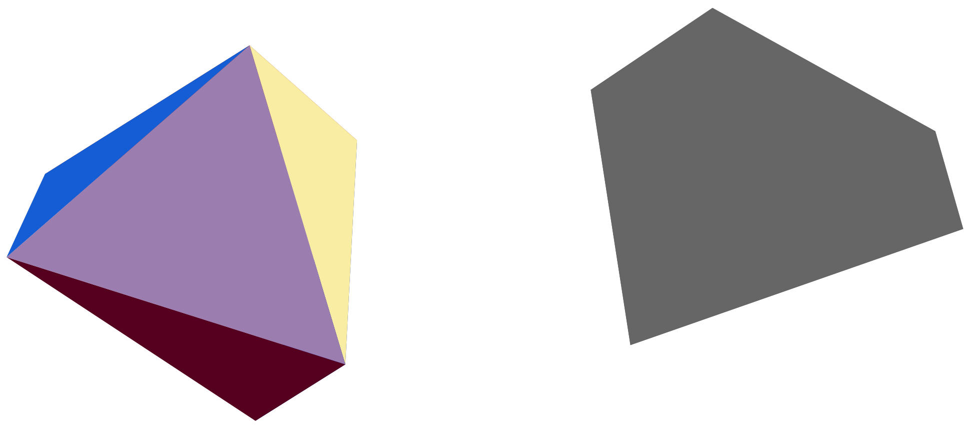

Figure 1: An example of a random polyhedral solid (left) and one of its projections (right).

optics with its ray tracing algorithms or Monte Carlo routines can now be applied to almost any reasonable geometry. These results, of course, can only be used if x is larger than about 100. It is one of our goals to extensively study if there exists an overlapping region for x where wave optical methods can still feasibly be used subject to limitations imposed by computers and where geometric optics already reasonably well approximates the exact results.

The environmental particles and those collected by stratospheric flights seem to appear in almost infinite number of various shapes. Therefore, it would be a fortune if at least a reasonable fraction of these particles could be divided in a small number of basic classes whose light scattering properties could then be calculated. In the present paper we propose one such classification in which we divide particles in only three classes: polyhedral solids, stochastic solids and stochastic aggregates. In the class of polyhedral solids all crystal shapes form just a subclass. For instance, sand particles seem to fit this class nicely. Stochastic solids are particles without any sharp edges. Typical examples of this class are some cosmic dust particles, atmospheric particulates as flyash and products by cement plants. Stochastic aggregates of smaller particles form a class in which most of the cosmic dust particles seem to fall. Examples of these scattering geometries are given in Figs. 1–3.

Our experimental setup (see Volten *et al.* [1]) is well suited to carry out controlled laboratory measurements of some members of the classes given above. It is our purpose both to measure and calculate all the elements in the 4×4 Mueller matrix to provide almost a unique comparison with measurements and theory.

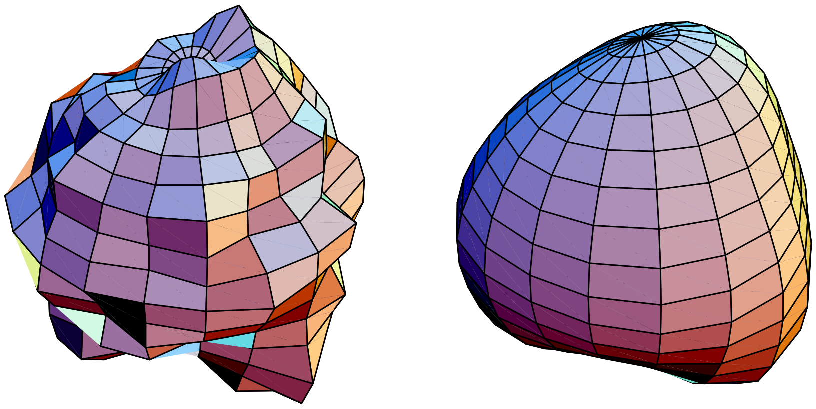

Figure 2: Two examples of stochastically rough particles.

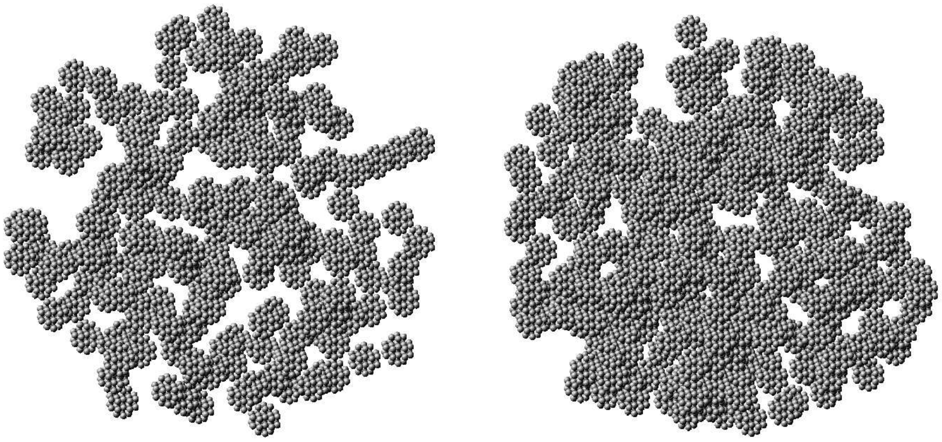

Figure 3: Stochastic aggregates of 200×32 (left) and 300×32 (right) dipoles.

2 Models for polyhedral and stochastically rough particles

2.1 Polyhedral particles

Muinonen *et al.* [2] have described how light scattering by various crystal shapes can be calculated using the geometric optics approximation. These calculations were further supplemented by adopting a first order physical optics correction based on the scalar Kirchhoff integral equation. This is an essential ingredient to smoothen otherwise infinitely narrow peaks in the forward and backward directions and also at the corner and trough reflections. The total number of rays used for any given crystal type was 10^6. The estimated error in results was less than 5 %.

Later Muinonen further improved the ray tracing code and introduced a concept of a modified Kirchhoff approximation (MKA) [3]. This was done with the vector Kirchhoff integral. It was estimated by Muinonen that the MKA produced reasonably accurate results for the size parameter as low as 25. This is, however, something which must be checked either by direct laboratory measurements or by rigorous wave optical codes such as the DDA or the T-matrix solution. Unfortunately, the T-matrix method is not well suited for particles with sharp edges because a very large number of spherical harmonics terms are needed to specify the surface. The DDA seems to work much better for such particles. Indeed, the code developed by Lumme and Rahola [4] is capable to handle particles with the size parameter as high as 20. Therefore, we are almost able to calculate light scattering in the whole size parameter range from the smallest Rayleigh particles up to really large particles provided that the MKA really works for the intermediate size range.

Kuik *et al.* [6] measured the Mueller matrix elements of some irregularly shaped quartz (SiO_2) particles with an effective radius of about 15μm. We plan to continue this work and concentrate on somewhat smaller SiO_2 particles. It is also our idea to study typical shapes of these particles in a quantitative sense. The particles measured by Kuik *et al.* seem to indicate that a reasonable approximation for a typical shape might be a random octahedron. We model this by choosing six random points on the surface of a sphere. Four of these points are located on the perimeter of the equatorial circle of the sphere, each in its own quadrant. Then one random point is chosen in the polar angle interval $0°$ to $45°$ and another one in the interval $135°$ to $180°$. An example of such an octahedral particle is shown in Fig. 1 together with its projection from one direction.

The existing ray tracing code by Muinonen *et al.* [2] to calculate light scattering by crystal shapes can easily be generalized to the random octahedral geometry. Results together with actual laboratory measurements will be published later.

2.2 Stochastically rough particles

Muinonen [7] described a stochastic particle shape by the multivariate lognormal probability density where the autocorrelation function for the logarithm of the radius is given by

$$D(\gamma) = \exp\left(-\frac{1}{2}\frac{\sin^2\gamma/2}{\sin^2\gamma_0/2}\right). \tag{1}$$

Here γ is the angular distance between any two given directions and γ_0 is the correlation angle. It is straightforward to expand Equation (1) in Legendre series

$$\begin{aligned} D(\gamma) &= \sum_{l=1}^{\infty} d_l P_l(\cos\gamma), \\ d_l &= (2l+1)\exp(-\eta) i_l(\eta), \\ \eta &= (2\sin^2\gamma_0/2)^{-1}, \end{aligned} \tag{2}$$

where i_l is the modified Bessel function. We then expand the logarithm of the particle radius in spherical harmonics as

$$u(\theta,\phi) = \sum_{l=1}^{\infty}\sum_{m=-l}^{l} P_l^m(\cos\theta)(a_{lm}\cos m\phi + b_{lm}\sin m\phi), \tag{3}$$

where a_{lm} and b_{lm} are independent Gaussian random variables with zero means and standard deviations σ_{lm}. These are related to d_l in Eq. (2) by

$$\begin{aligned} \sigma_{lm}^2 &= \frac{2\pi}{2l+1}\beta^2 d_l, \\ \beta^2 &= \log(1+\sigma_a^2/a^2), \end{aligned} \tag{4}$$

where σ_a^2 is the variance of the radius a of the original unperturbed sphere. Two examples of the stochastically rough particles are given in Fig. 2.

In our ray tracing approximation four Stokes parameters are related to every ray. At the boundary, reflection and refraction take place according to Snell's law and Fresnel's coefficients. The scattering phase matrix can be divided

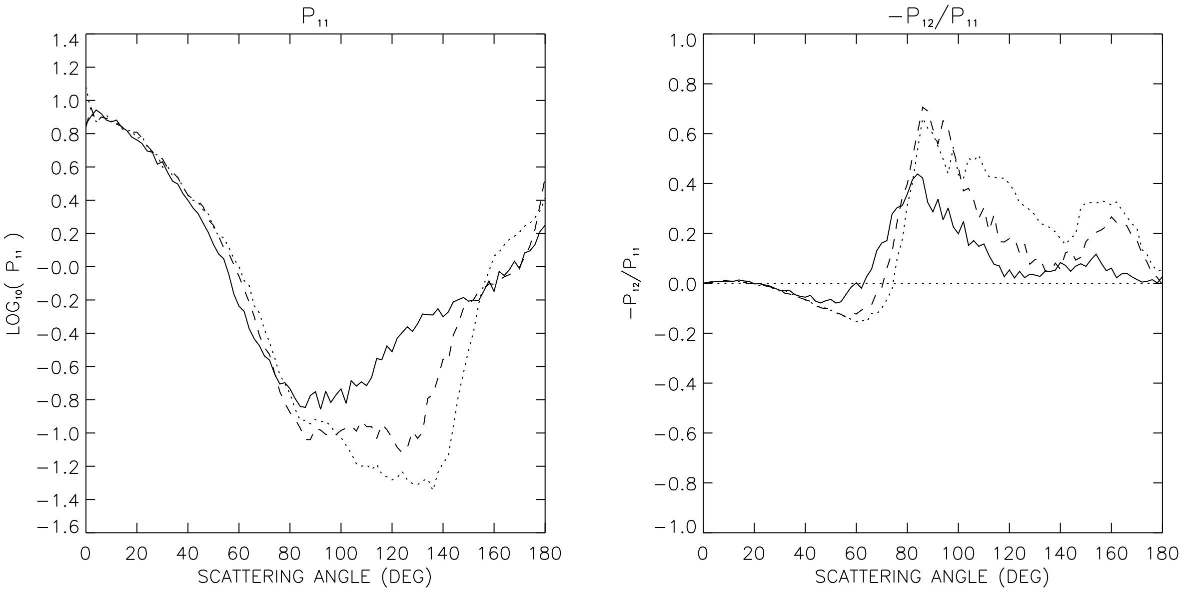

Figure 4: The scattering matrix elements P_{11} and P_{12} for three stochastically rough particles. The correlation angle of the radius is $\gamma_0 = 30°$, $60°$ and $90°$ (solid, dashed and dotted lines, respectively).

into the diffraction and geometric optics parts by

$$P = \frac{1}{2\sigma_{sca}} \left(\sigma_{ext} P^D + (2\sigma_{sca} - \sigma_{ext}) P^G \right), \tag{5}$$

where σ_{sca} and σ_{ext} stand for the scattering and extinction cross sections, respectively. The forward diffraction part of Eq. (5) can be written as

$$\begin{aligned} P^D &= \frac{k^2}{4\pi G} |v(\theta,\phi)|^2 (1+\cos\theta)^2 I, \\ v(\theta,\phi) &= \int_0^{2\pi} d\phi' \int_0^{r(\phi')} dr' r' \exp(-ikr' \sin\theta \cos(\phi - \phi')), \end{aligned} \tag{6}$$

where k is the wavenumber, G is the cross-sectional area, I is the 4×4 identity matrix and $r(\phi')$ the silhouette of the particle.

In the computations the cutoff value for the flux was set at 10^{-4} and a maximum of ten internal reflections were included in the analysis. A total of $2.5 \cdot 10^4$ rays were traced in each case. Results for the matrix elements P_{11} and P_{12} are shown in Fig. 4. A more detailed description of our procedure and results will be published later.

3 Light scattering by fluffy aggregates

Lumme and Rahola [8] adopted the DDA to calculate light scattering by fluffy particles. They modeled the geometry with equal sized spheres (32 dipoles, size parameter 1.2) were randomly placed in 100, 200 and 300 lattice sites inside a pseudosphere [5] of 1064 possible sites. The resulting size parameter x_1 of the aggregate can be calculated from [8]

$$x_1 = x_0 \left(32\frac{6}{\pi}\right)^{1/3} \left(1064\frac{6}{\pi}\right)^{1/3}, \tag{7}$$

where x_0 is the size parameter of a single dipole, assumed to be 0.3. Thus we have $x_1 = 14.9$. The three choices for the number of the constituent spheres results in packing densities of 0.1, 0.2 and 0.3. The geometry of the two densest cases are shown in Fig. 3.

3.1 The discrete-dipole approximation

In the DDA a particle is modeled as a group of interacting electric dipoles. The dipole moments $\mathbf{p}_i$ and thus the electric fields $\mathbf{E}_i$ radiated by the dipoles are unknown. The total electric field at the dipole positions is determined by solving the system of linear equations for $\mathbf{E}_i$ (see e.g. [4]),

$$\mathbf{E}_i = \mathbf{E}_i^0 + k^3\alpha \sum_{j\neq i}^{N_{dip}} \mathbf{T}_{ij} \cdot \mathbf{E}_j, \quad i = 1, \ldots, N_{dip}, \tag{8}$$

where $\mathbf{E}_i^0$ is the incident field at a position $\mathbf{r}_i$, k is the wave number and α is the polarizability of a dipole. The dipole interaction tensor (or the Green dyadic) is given by

$$\begin{aligned} \mathbf{T}_{ij} &= \frac{e^{i\rho_{ij}}}{\rho_{ij}^3} \left[(\rho_{ij}^2 + i\rho_{ij} - 1)\mathbf{1} + (3 - 3i\rho_{ij} - \rho_{ij}^2)\hat{\mathbf{r}}_{ij}\hat{\mathbf{r}}_{ij}\right], \\ \rho_{ij} &= k|\mathbf{r}_i - \mathbf{r}_j|, \end{aligned} \tag{9}$$

where $\mathbf{1}$ is the unit dyadic and $\hat{\mathbf{r}}_{ij}\hat{\mathbf{r}}_{ij}$ is a dyadic formed from the interdipole unit vectors $\hat{\mathbf{r}}_{ij} = (\mathbf{r}_i - \mathbf{r}_j)/|\mathbf{r}_i - \mathbf{r}_j|$.

The bottleneck in the DDA is the question of the correct expression for $k^3\alpha$ because we are dealing with finite sized spherical dipoles which do not fill a particle completely. Lumme and Rahola [4] assumed in accordance with Mie theory

$$k^3\alpha = x_0^3\frac{m_d^2 - 1}{m_d^2 + 2} + \frac{3}{5}x_0^5\frac{(m_d^2 - 2)(m_d^2 - 1)}{(m_d^2 + 2)^2} + i\frac{2}{3}x_0^6\left(\frac{m_d^2 - 1}{m_d^2 + 2}\right)^2, \tag{10}$$

where x_0 is the size parameter of a dipole and m_d its refractive index. To relate m_d to the refractive index m of the real material, we are forced to use some effective medium theory. Lumme and Rahola adopted a semi-empirical model by Sihvola [9] which states that

$$\frac{m^2-1}{m^2+2+\nu(m^2-1)} = f\frac{m_d^2-1}{m_d^2+2+\nu(m_d^2-1)}, \tag{11}$$

where f is the volume fraction (packing density) of a particle modeled with densely packed dipoles. A natural value for f is the ratio of the volume of a sphere to that of a circumscribed cube, i.e. $\pi/6$. Sihvola showed that Eq. (11) agrees with the classical Maxwell Garnett theory if $\nu = 0$, with Bruggeman theory if $\nu = 1$, Polder-van Sauter theory if $\nu = 2$, and finally with the quasi-crystalline approximation with coherent potential if $\nu = 3$. We have allowed the parameter ν to take any values, either positive or negative, which best approximate Mie theory. Almost independently of m or x_0 we have found that $-0.3 \leq \nu \leq 0$.

To model an icy particle we set $m = 1.29 + 0.008i$ which corresponds, Eq. (11), to $m_d = 1.6 + 0.02i$. For a silicate particle we assume $m = 1.57 + 0.012i$ which in turn translates to $m_d = 2.5 + 0.05i$. In both cases we have assumed $\nu = -0.1$.

3.2 Computations

Computationally the most intensive part of the DDA calculations is the solution of dense systems of linear equations, where the coefficient matrix is complex symmetric. Previously the linear systems were solved by the conjugate gradient method applied to the normal equations. We have utilized a new iterative method called QMR (quasi-minimal residual method), see Freund [10], which converges three to four times faster than the conjugate gradient method [11].

In the application of iterative methods to the DDA problem, almost all time is spent in the computation of the product of the coefficient matrix with a given vector. We have applied two fast methods to compute this matrix-vector product, the FFT method and the Fast Multipole Method (FMM). If the dipoles sit on a regular lattice, the matrix-vector product reduces to a 3D convolution, which can efficiently be computed with the FFT algorithm (see Goodman, Draine, and Flatau [12]). We have been able to calculate the scattering by over 100 000 dipoles with the FFT algorithm. This took a few hours of CPU time on a Convex C3840. In this paper, the computation of scattering by the particle with 9600 dipoles took some 10 minutes to compute for a single orientation.

In the FMM algorithm the matrix-vector product is computed by truncated potential expansions of the dipole potentials, see Greengard and Rokhlin [13]. The development of the FMM algorithm is still under way. Our preliminary experience shows that the FMM can be competitive with the FFT algorithm especially when the computational geometry is sparse, such as a diffusion-limited aggregation cluster [14].

3.3 Results

The DDA computations are based on several hundred orientation averages of the geometries shown in Fig. 3. Our criterion for the proper number of those averages is such that polarization both in the forward and backward directions must be zero with less than 1 %.

It is interesting to compare our results to calculations based on pure geometric optics with the same geometries. We have, therefore, carried out Monte Carlo simulations in spherical geometry with three different radial optical thicknesses of 0.5, 1.0 and 1.5, which correspond to our three packing densities. The number of photons used in MC simulations was $5 \cdot 10^4$ which turned out to be sufficient. In our computations we used the full 4×4 scattering matrix for which the elements were obtained from the single particle ($x = 1.2$) scattering. One example of our DDA computations together with MC simulations is given in Fig. 5. As we can see from the figure, the wave and geometric optical calculations agree surprisingly well, although we are dealing with small clusters. Two distinctive differences are, however, obvious. The diffraction part of P_{11} cannot, of course, be explained by geometric optics. Also the negative part of linear polarization cannot be reproduced. Actually all our efforts to produce negative polarization with geometric optics in any reasonable geometry have always badly failed.

4 Laboratory measurements

To be able to compare the computed results with laboratory measurements, we make use of measurements performed with an experimental setup from the Free University in Amsterdam (see Volten *et al.*, this proceedings [1]). In this experimental setup a HeNe laser is employed as a polarized light source ($\lambda =$ 632.8 nm). The light is scattered by randomly oriented aerosols or hydrosols. All relevant scattering matrix elements are obtained from four or five separate

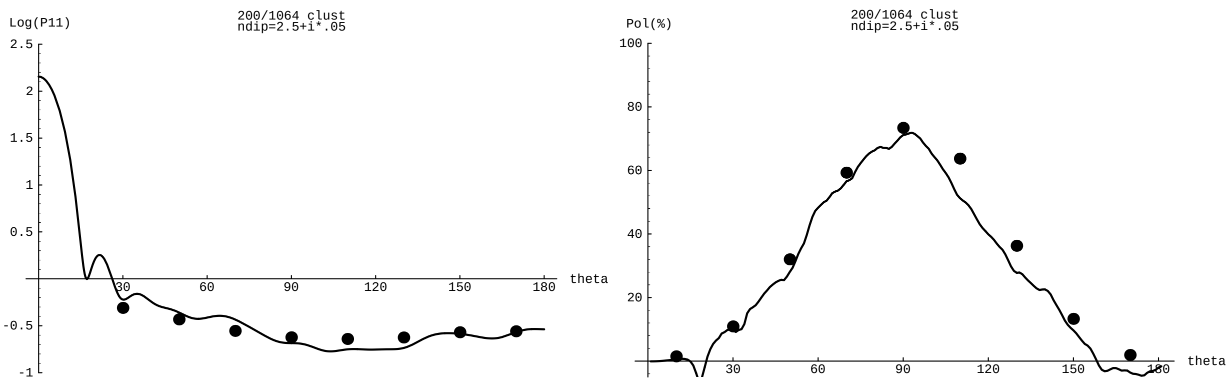

Figure 5: The single particle phase function P_{11} and the linear polarization for the stochastic aggregate of 200×32 dipoles (see left half of Fig. 3). DDA results are given by the solid line and the results by Monte Carlo method are given by dots.

Figure 6: A typical SiO_2 particle.

measurement runs, by using a polarization modulation technique.

With this setup we plan to do some measurements that are indicative of the scattering behaviour of polyhedral solids, namely SiO_2 particles, and certain types of algae which can be described as stochastic aggregates.

A typical SiO_2 particle (see Fig. 6) has a chunky irregular shape and is not very elongated. The average aspect ratio of these particles is 1.7 (ref.?). Their refractive index is $1.53 + 0.0045i$. The size of sand aerosols found in nature range from 0.02 μm up to more than 100 μm. However, only particles in the size range between 0.1 μm and 5 μm are transported over large regions. In our experiment we will use aerosols with a mean radius of a few μm. These particles are not only most interesting for climate studies but also are in a convenient size range for the calculations.

Obtaining the scattering properties of algae is important for the interpretation of e.g. remote sensing data. Algae with a size range of typically a few μm are

Figure 7: A collection of algae.

very common. Although a general description of the geometry of the algae cannot be given, since the differences between types of algae can be very large, it seems reasonable to describe certain types of algae in terms of stochastic aggregates, e.g. as a sphere or spheriod containing several pigment or gaseous bubbles (see Fig. 7).

5 Conclusions

In this paper we have shown how to compute light scattering by three particle groups: polyhedral particles, stochastically rough particles and fluffy aggregates. We have shown new computational results of the last two cases. In the near future we plan to measure in laboratory sand particles (SiO_2) and some algae. Comparison between these measurements and our models will provide new insight of light scattering by various complicated structures.

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