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Abstract. — A numerical method is described which is able to calculate the scattering of polarized light by a large assembly of identical spherical particles, of complex refractive index, such as a colloid or aerosol aggregate. The method takes care of multiple scattering inside the aggregate, except that the criterium for the Rayleigh approximation must be satisfied for the individual particles. It is applied to computer-generated aggregates containing up to 512 particles with various fractal dimensions. The scattered intensities for various polarization directions are studied as a function of the scattering angle and size of the aggregate.

1. Introduction.

It is of fundamental importance to understand the scattering of light by an assembly of particles such as colloid or aerosol aggregates. This is of practical interest in many areas of applied science and technology. For example, in the case of aerosols, this study could provide a better knowledge of the optical properties of smoky atmospheres.

Since the discovery of the fractal structure of aerosol [1] and colloid [2] aggregates, there have been several attempts to study light scattering by fractal objects. The most simple scalar-field scattering theory, which neglects multiple scattering, gives a scattering intensity which is simply the Fourier transform of the pair correlation function between particles in the structure [3]. As a result, for fractal structures, the scattered intensity varies as \( q^{-D} \), where \( D \) is the fractal dimension, in the limit where the modulus of the scattering vector \( q = (4 \pi / \lambda) \sin (\theta / 2) \) lies between the two natural cut-off \( q_{\text{min}} = L^{-1} \) and \( q_{\text{max}} = a^{-1} \), where \( L \) is the size of the aggregate and \( a \) the radius of the individual particles [4]. This result has led to numerous experimental investigations of the fractal dimension of aggregates via scattering techniques [2, 5]. However, in the case of light scattering there are severe limitations in neglecting multiple scattering. First, the size of a typical aerosol aggregate can be of the order of the wavelength of visible light. In this case, for quite opaque aggregates (more precisely for large fractal dimensions [6]), one can expect non negligible multiple scattering contributions to the scattered intensity. Second, light is not a scalar but a vector field, and if one performs scattering experiments with polarized light, one can expect that some geometrical extinctions are removed when multiple scattering is allowed.

In this paper we describe a numerical method able to calculate the scattered intensity in any direction and polarization by an assembly of identical spherical particles enlightened by a monochromatic planar incident wave with a given polarization. The only ingredients are the wavelength of the incident light, the radius of the individual spherical particles, their optical characteristics, which can be summarized by...
a complex refractive index, \( m = m' + i m'' \), and their positions in space (i.e. a list of coordinates). Our method takes care of multiple scattering. The only important limitation is that the Rayleigh approximation must be verified for the particles, i.e. \( ka = 0.3 \), where \( k = 2 \pi / \lambda \) is the modulus of the wave vector of the incident beam and \( a \) the radius of the particles. Note that such a condition does not exclude large \( kL \) values, where \( L \) is the size of the aggregate. Thus, for sufficiently big aggregates, multiple scattering effects are supposed to be fully taken into account. Our method has been applied to several computer-generated sets of different fractal dimensions and a systematic study of the behavior of the scattered intensity as a function of the size of the aggregates has been performed. We give the main principles of our method in part 2, the results in part 3 and a discussion in part 4.

2. Principles of the numerical calculation.

Let us consider an aggregate containing \( N \) identical spherical particles, of radius \( a \), centred at positions \( r_i, i = 1, 2, ..., N \) in the regular three dimensional space. One assumes that the aggregate is connected in the sense that each particle contacts at least one other particle, the distance between the center of two nearest neighboring particles being strictly equal to the particle diameter, \( 2a \). This aggregate is enlightened with an incident monochromatic plane wave of wavevector \( \mathbf{k} \), whose modulus is \( k = 2 \pi / \lambda \) and whose direction is called \( z \). The incident electric field at position \( \mathbf{r} = (x, y, z) \) is thus given by:

\[
E_{\text{inc}} = E_0 \exp(i \mathbf{k} \cdot \mathbf{r}) = E_0 \exp(i k z) .
\]

The orientation of the field \( E_0 \) defines the polarization of the incident light.

Each particle, \( i \), feels the field of the incident beam as well as the fields scattered by all the other particles, \( j \neq i \). To calculate these scattered fields, one assumes that each homogeneous spherical particle behaves as an oscillatory electrical dipole \( \mathbf{p}_i \), proportional to the electrical field inside the sphere, denoted \( \mathbf{E}_i \):

\[
\mathbf{p}_i = \frac{4 \pi}{3} \varepsilon_0 a^3 \alpha_i \mathbf{E}_i
\]

where \( \alpha_i \) is, in general, a tensor characterizing the polarizability of the particle. Practically, in all our calculations, the polarizability tensor has been considered as constant and diagonal, simply related to the complex refractive index, \( m \), of the particles through the formula:

\[
\alpha_i = \varepsilon - 1 = m^2 - 1 = m'^2 - m''^2 - 1 + 2 i m' m''.
\]

One could have introduced some non-diagonal terms to take care of the local anisotropy due to the neighboring particles, however we have checked that this modification does not change the main results presented below.

This induced dipole creates a scattered wave in the whole space. As well known in the standard Rayleigh theory, this scattered wave becomes purely spherical at large distances. However, at intermediate distances, one must introduce a tensor to calculate the electrical field scattered by a particle \( i \) and created in the vicinity of, but outside, another particle \( j \):

\[
E_{i,\text{ext}} = (ik^3/4 \pi \varepsilon_0) T_{ij} \mathbf{p}_i
\]

In [4], we have followed the notations of Jones [7] who performed quite similar calculations, but on aggregates of very limited number of particles (up to 10). \( T_{ij} \) is in general a dimensionless tensor which depends on \( k, r_i \) and \( r_j \). In the limit of large distances between particles, i.e. when \( kr_{ij} \gg 1 \), where \( r_{ij} \) is the modulus of the vector \( \mathbf{r}_{ij} = \mathbf{r}_j - \mathbf{r}_i \), this tensor must reduce to the following operator:

\[
T_{ij} = \exp(ikr_{ij})/(ikr_{ij}) \mathbf{u}_i \times \mathbf{u}_j \times
\]

where \( \mathbf{u}_j = \mathbf{r}_{ij}/r_{ij} \) and where \( \times \) denotes the vectorial product. This formula corresponds to the leading term of an expansion in \( 1/r_{ij} \) of the general tensor, which is in principle fully determined by the Mie theory [8]. In our calculations, as in the old ones by Jones [7], we have taken an expansion up to the second order in \( 1/r_{ij} \), i.e. up to the dipolar terms. This takes care only partially of the errors made when considering neighboring particles. An attempt to take into account the full Mie theory is under progress [9]. The detailed expression of our \( T_{ij} \) operator is given in Appendix.

Then, using the relation between the field inside and outside an homogeneous sphere:

\[
E_j = (\alpha_j/3 + 1) E_j
\]

one obtains, by combining (2), (4) and (6), the following self consistent set of equations for the fields \( E_j \):

\[
(\alpha_j/3 + 1) E_j = E_0 \exp(i k z) + i (ka)^3/9 \sum_{i \neq j} T_{ij} \alpha_i E_i .
\]

As soon as this system is solved, the total scattered electrical field can be determined everywhere in space and, in particular, on a detector, through the formula:

\[
E_d = i (ka)^3/9 \sum_i T_{id} \alpha_i E_i .
\]

If the detector is placed far away from the aggregate
in a given direction defined by a unit vector \( \mathbf{v} \), (8) reduces to:

\[
E_d \propto k^2 a^3 \mathbf{v} \times \mathbf{v} \times \sum_i \alpha_i E_i \exp(-i k_d \cdot r_i)
\]

where \( k_d = k \mathbf{v} \).

The only delicate technical problem remains to solve the system of equations (7) for the fields. One could have used standard library subroutines to invert a \( 3N \times 3N \) matrix, as it was done in the paper by Jones [7], but due to the cost of memory, this would limitate our calculations to aggregates of no more than one hundred particles. We have preferred to use an iterative method, which was already proposed in the past by Ravey [10]. In this method we start from:

\[
E_i^{(0)} = (\alpha_i/3 + 1)^{-1} E_0 \exp(i k z_i) = (\alpha_i/3 + 1)^{-1} E_0 \exp(i k \cdot r_i)
\]

and we calculate the \( n \)th approximant, \( E_i^{(n)} \), through the recursion formula:

\[
E_i^{(n+1)} = (\alpha_i/3 + 1)^{-1} [E_0 \exp(i k z_i) + i (k a)^3/9 \sum_{\omega_j} T_{ij} \alpha_j E_j^{(n)}] .
\]

The advantage is that now it is only necessary to store in memory vectors of length \( 3N \), but the inconvenient is that the elements of the tensor must be recalculated at each iteration. In practice, we have found that this iterative method is quite quickly converging. For \( k a = 0.1 \), it is only necessary to go up to orders 5 or 6 to obtain relative errors lower than 1\% on the fields. For larger values of \( k a \), one must go further. In principle, this method allows us to treat aggregates of up to some thousands of particles. In practice, to have a good statistics, we limitated ourselves to aggregates up to \( N = 512 \) particles. Before using our method on computer generated fractal aggregates, we have tested it on an alignment of ten sphere with various orientations, and we have recovered the results of Jones [7] within a good accuracy.

3. Application to fractal aggregates.

In order to clearly show the influence of the fractal dimension on the scattered intensity, we have built different clusters of increasing fractal dimensions, using different aggregation models [11]. Typical clusters of 2 048 particles, corresponding to the models used, are shown in figure 1. The common feature of all these aggregates is that they have been built off-lattice to avoid strong angular correlations. They are labelled from (1) to (4) according to increasing fractal dimension.

Cluster (1) has been built using the particle-cluster « tip-to-tip » model [12]. In this model, particles are added, one after another, on the most external particle in successively randomly chosen space direction. Eventhough the same kind of random screening effect occurs as in the Witten-Sander model [13], this aggregate has a fractal dimension trivially equal to 1. In practice there are some finite-size effects, and if one calculates on effective fractal dimension for finite clusters, it would be larger than 1 an tending to 1 when increasing the cluster size.

Cluster (2) has been built using the hierarchical cluster-cluster « tip-to-tip » model (12). In this model, clusters of equal number of particles are iteratively stuck together after having been randomly rotated in space, placed far apart from each other in a randomly chosen direction, and finally joined in contact on their neighboring particles. The fractal dimension of such cluster is \( D = 1.5 \), smaller than those obtained for clusters built with the brownian cluster-cluster model [14].

Cluster (3) has been built using the hierarchical cluster-cluster model with random linear trajectories [15]. In this model, clusters of equal number of particles are iteratively stuck together after moving along a random straight line in space. The fractal dimension is \( D = 1.9 \), slightly larger than those obtained for clusters built with the brownian cluster-cluster model [14].

Finally, cluster (4) has been built using the particle-cluster model with random linear trajectories [16]. It is known that this model leads to compact clusters of fractal dimension equal to the dimension of space, here \( D = 3 \). Here also there are some finite-size effects, and if one calculates an effective fractal dimension for finite clusters, it would be smaller than 3 and tending to 3 when increasing the cluster size.

In each case, for clusters of sizes \( N = 8, 16, 32, 64, 128, 256, 512 \), we have calculated the scattered intensity in a direction making an angle \( \theta \) with the direction of the incident beam. The complex index of the spheres has been taken to be \( m = 1.6 - 0.8i \), the same value as in Jones [7]. After having tested that the \( m \) values does not influence the generality of the qualitative results presented here, we do not present any systematic study as a function of \( m \). To show the effect of the product \( k a \), we have chosen only two values for this parameter, \( k a = 0.1 \), which satisfies the Rayleigh criterium for the particles, and \( k a = 0.5 \) which has been chosen deliberately slightly too large to better show the effect of increasing multiple scattering effects. The intensity has been averaged over 50 different orientations of the cluster, randomly chosen in space, so that the results should not depend on the azimuthal orientation of the detector. This would simulate a real scattering experiment in which the clusters are randomly
Typical examples of aggregates on which the light scattering intensities have been calculated. These aggregates have been built with the same number of particles \(N = 2048\). The grey tones indicate the deepness. The fractal dimensions are \(D = 1, 1.5, 1.9, 3\) when going from case (1) to case (4), respectively. The aggregates are represented with the same apparent size, so that the change in the fractal dimension can be seen on the variation of the apparent particle diameter.

Oriented in space. However, in real experiment there is a given size distribution of clusters which is not reproduced here. In the case of the cluster-cluster models (2) and (3), we were also able to make averages over all the independent cluster configurations of \(N\) particles leading to the configuration of 2048 particles shown in figure 1 (i.e. 4 configurations for \(N = 512\), 8 for \(N = 256\), etc...). This allowed us to test the average on the 50 orientations of the same cluster. We observed that, due to the random character of the clusters, this average was quite efficient even for the very elongated structure obtained in case (1). The intensity is noted \(I_{AB}\), where the indices A and B refer to the orientations of the polarizer and the analyzer, respectively. These indices are denoted H, when the polarization is in the incident plane (i.e. the plane containing both the incident and the scattered directions), and V, when the polarization is perpendicular to this plane.

In figure 2, we give typical results for the curves
The natural logarithm of \( I_{HH}, I_{VH}, I_{HV}, I_{VV} \), plotted as a function of the scattering angle \( \theta \), in the case (2) \((D = 1.5)\), for aggregates containing \( N = 512 \) particles. Cases (a) and (b) correspond to \( ka = 0.1 \) and \( ka = 0.5 \) respectively.

In \( (I_{HH}), \ln (I_{HV}) \) versus \( \theta \), in the case (2) \((D = 1.5)\) and for \( N = 512 \). Case (a) corresponds to \( ka = 0.1 \) and case (b) to \( ka = 0.5 \). Some qualitative features are common to such curves. While \( I_{VV} \) decreases monotonically from \( \theta = 0 \) to \( \theta = \pi \), \( I_{HH} \), which is equal to \( I_{VV} \) in these two limits for obvious symmetry reasons, goes through a minimum near \( \theta = \pi/2 \). This minimum corresponds to the geometrical extinction predicted by the simple theory without multiple scattering. This is due to the fact that, when observing at \( \theta = \pi/2 \), with an horizontal incident polarization, the field of the incident light is in the direction of observation. Also the cross intensities \( I_{VH} \) and \( I_{HV} \) are very small, do not depend too much on \( \theta \), and are of the same order of magnitude than the minimal value of \( I_{HH} \). The effect of increasing \( ka \) is to decrease the backscattering intensities \( I_{VH}(\pi) = I_{HH}(\pi) \). Similar curves have been obtained with the other aggregates and for different sizes. In the following figures, we systematically analyse their main characteristics as a function of the aggregate size.

In figures 3(a) and 3(b), we report the results for \( I_{VV} = I_{HH} \) for \( \theta = 0 \) and \( \theta = \pi \). The logarithm of the intensity is plotted as a function of the logarithm of \( N \), the number of particles in the aggregate. In 3(a), all the results corresponding to \( ka = 0.1 \) are reported, while in 3(b), the preceeding data are compared with those corresponding to \( ka = 0.5 \) only for \( D = 1.5 \) and \( D = 1.9 \). While all the direct scattering intensities are grouped on the same straightline independent on the fractal dimension, with a slope equal to 2, the blackscattering ones depend on \( D \) and show a crossover from a slope of order 2 for small sizes to a smaller slope, of order 1, for larger sizes. As observed on figure 3(b) the small size effect disappears for larger \( ka \) values. Also increasing \( ka \) increases slightly the direct scattering intensity but does not change the slope.

In figures 4(a) and 4(b), we report the results for \( I_{VH} = I_{HV} \) for \( \theta = 0 \) and \( \theta = \pi \) plotted as a function of the natural logarithm of \( N \). (a) corresponds to \( ka = 0.1 \) while in (b), the curves for \( ka = 0.1 \) and \( ka = 0.5 \) are compared in the cases \( D = 1.5 \) and \( D = 1.9 \).
natural logarithm of the depolarisation factor, i.e. \( \ln (I_{VV} / I_{VH}) \), for direct scattering \((\theta = 0)\) and backscattering \((\theta = \pi)\). Note that these results should correspond to a very simple experiment (rotating a polarizer). While there is not too much difference between direct and back scattering for large fractal dimensions, one observe a difference which clearly increases with \(N\), for smaller fractal dimensions.

At last, to test if the \(q^{-D}\) behavior is recovered with our method we report in figure 6 the log-log plots of \(I_{VV}\) versus \(q = 2k \sin (\theta /2)\) for clusters of 512 particles.

4. Discussion of the results.

The main features concerning \(I_{VV}\) and \(I_{HH}\) (Fig. 3) can be understood using the scalar theory without multiple scattering [4]. This theory can be simply recovered using the zero-th order expressions for the fields \(E_i\) in the expression of the total scattered field on the detector given by formula (8). Forgetting the polarizability factors, this gives:

\[
E_d \propto k^2 a^3 \mathbf{v} \times \mathbf{v} \times E_0 \sum_i \exp(-i q \cdot r_i)
\]
where \( q = k_d - k \) is the scattering vector whose modulus is \( q = 4\pi\sin(\theta/2)/\lambda \). Taking the square of the modulus of the field to get the intensity, we find for \( I_{vv} \), for which the geometrical factor \( \mathbf{v} \times \mathbf{v} \times \mathbf{E}_0 \) does not depend on \( \theta \):

\[
I_{vv} \propto \sum_i \sum_j \exp(i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)).
\]

(13)

Transformed into an integral this double sum appears to be the Fourier transform of the correlation function, \( \rho(r) \), between centers of two particles in the aggregate. Let us assume that \( \rho(r) \) is spherically symmetrical and simply described by a power law \( r^{-(3-D)} \), multiplied by some exponential cut-off function, to take care of the finite size of the aggregate:

\[
\rho(r) \propto (N/G(D)) r^{-(3-D)} \exp(-r/R).
\]

(14)

where \( R \) is a characteristic radius for the aggregate, slightly larger than its radius of gyration, and where the gamma function appears for reasons of normalization.

The scattering intensity can be calculated by Fourier transforming to get:

\[
I(q) \propto \left[N^2/(D-1)\right] (qR)^{-(D-1)} \times [1 + (qR)^{-2}]^{(D-1)/2} \sin [(D-1)\tan^{-1}(qR)].
\]

(15)

In the limit of small \( qR \) values, (15) reduces to:

\[
I(q) \propto N^2 q^{-2(D-1)} \sin ((D-1)\pi/2).
\]

(16)

independently on \( D \), as it could have been directly seen using the original double sum. In this limit, all particles diffuse coherently. This explains why \( I(0) \) scales like \( N^2 \), independently on \( D \), and also why for \( \theta = \pi \), and small sizes, \( I(q) \) scales as \( I(0) \). For large values of \( qR \) instead, (15) reduces to:

\[
I(q) \propto N (q^{-D}/(D-1)) \sin ((D-1)\pi/2).
\]

(17)

This explains why \( I(q) \) scales like \( N \) for \( \theta = \pi \) and for sufficiently large sizes. The crossover between \( N^2 \) and \( N \) behaviors occurs for \( qR \) of order unity, i.e. in the case \( \theta = \pi \) for \( R \) of order \( \lambda/(4\pi) = k^{-1}/2 \). This explains why the crossover is located at smaller sizes for larger ka values.

The results for \( I_{vH} \) and \( I_{HV} \) (Fig. 4) are more delicate to explain. In this case the above zero-th order term becomes strictly zero due to pure geometrical reasons. Going then up to the first order term, and forgetting again all polarizability factors, one gets:

\[
E_d \propto k^5 a^6 \mathbf{v} \times \mathbf{v} \times \sum_i \sum_j T_{ij} \mathbf{E}_0 \exp(i(\mathbf{k} \cdot \mathbf{r}_i - k_d \mathbf{r}_j)).
\]

(18)

To obtain the asymptotic large-\( N \) regime one can replace \( T_{ij} \) by its Rayleigh approximation in \( r_i^{-1} \). When there is no coherence effect, the double-sum in the expression of \( E_d \) behaves as \( N \langle r_i^{-1} \rangle \) in the same way as \( I_{vv} \) behaves as \( N \) when \( \theta \neq 0 \). Thus, for the large-\( N \) scaling of \( I_{vH} \), one expects:

\[
I \propto N^{2-2/D}.
\]

(19)

The slope \( 2-2/D \) has been indicated in figure 4. This slope is only approximately recovered in our numerical calculations however the same variation with \( D \) is obtained. We must note that there is no serious reason to trust such exponent since only the first order contribution has been taken into account.

At last, we must give some comment on the log-log plots of \( I_{vv} \) versus \( q \) of figure 6. The \( q^{-D} \) law is in principle valid between \( L^{-1} \) and \( a^{-1} \), where \( L \) is the size of the aggregate. However, here, since \( q \) is varied via \( \theta \), there is a geometrical upper cut-off which corresponds to \( \theta = \pi \), which is for \( q \) of order \( 2k = (2ka)^{-1} \). This explains why the range of the linear behavior is larger in case (b) \((ka = 0.5)\) than in case (a) \((ka = 0.1)\). Also, the range of validity is smaller for larger fractal dimensions, since for a given number of particles (here \( N = 512 \)), the ratio \( L/a \) decreases when \( D \) increases. This explains why we do not find any linear behavior for \( D = 1.9 \) and \( D = 3 \) in case (a). Considering this artifact and also the errors coming from the small size of our clusters, it is very difficult to conclude on the validity of the \( q^{-D} \) law in presence of multiple scattering.

5. Conclusion.

In this paper, we have presented a numerical method able to calculate the scattering of light by an assembly of identical spherical particles and we have applied it to computer-generated fractal aggregates. The scattered intensities with various directions of the polarizer and analyzer \( I_{vv}, I_{HH}, I_{vH}, I_{HV} \) have been calculated and systematically studied as a function of scattering angle, cluster size and fractal dimension. If the behaviors for \( I_{vv} \) and \( I_{HH} \) are well known and can be simply explained by the standard scattering theory, the results for the cross intensities \( I_{vH} \) and \( I_{HV} \) are new and can only be explained by invoking multiple scattering effects. Our results suggest very simple experiments, such as the determination of depolarization factors for direct and back scattering. Such experiments are under progress on aerosols [17]. Thus, the next step will be to fit the experimental results. A trivial result is that multiple scattering effects become much more important for larger values of the parameter \( ka \). We were limited in increasing this parameter here since we were
obliged to stay within the Rayleigh approximation for the individual particles. We hope that this constrain will be soon removed, since a method is under progress [9] which takes care of the full Mie theory. The effect of multiple scattering also increases as \( D \) becomes larger, this is consistent with the results of Berry's theoretical work [6]. To conclude, we must recall that \( D \) is certainly not the unique parameter able to characterize the physical properties of a fractal structure and it might be that another parameter, such as the spectral dimension, could enter the light-scattering results. However, in the loopless structures considered here, the spectral dimension varies monotonically with the fractal dimension and our calculations are not sufficient to test this idea.

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Appendix.
If the polar coordinates of \( r_{ij} \) are \( r, \theta, \varphi \), the tensor \( T_{ij} \) writes (8):

\[
\begin{array}{ccc}
  a & b & c \\
  a' & d & e \\
\end{array}
\]

with

\[
a = -h_0^{(1)}(kr) - h_2^{(1)}(kr) \times \left[ P_2(\cos \theta) - \cos(2\varphi)P_2^2(\cos \theta)/2 \right]
\]

\[
a' = a = -h_0^{(1)}(kr) - h_2^{(1)}(kr) \times \left[ P_2(\cos \theta) + \cos(2\varphi)P_2^2(\cos \theta)/2 \right]
\]

\[
b = h_2^{(1)}(kr) \sin(2\varphi)P_2^2(\cos \theta)/2
\]

\[
c = -h_2^{(1)}(kr) \cos(\varphi)P_2^1(\cos \theta)
\]

\[
d = -h_2^{(1)}(kr) \sin(\varphi)P_2^1(\cos \theta)
\]

and

\[
e = 2h_2^{(1)}(kr)P_2(\cos \theta)
\]

where the expressions for the spherical Bessel functions \( h_n^{(m)}(x) \) and the Legendre functions \( P_n(x) \) and \( P_n^{(m)}(x) \) can be found in standard tables [18].

References

[10] See, for example, Bohren, C. and Huffman, D., Absorption and scattering of light by small particles (Wiley Interscience) 1983.