Scattering of particles with inclusions. Modeling and inverse problem solution in the Rayleigh-Gans approximation

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Abstract. An analytic model for the scattering of a spherical particle with spherical inclusions has been proposed under the RG approximation. The model can be used without limitations to describe an X-ray scattering experiment. However, for light scattering several conditions must be fulfilled. Based on this model an inverse methodology is proposed to estimate the radii of host particle and inclusions, the number of inclusions and the Distance Distribution Functions (DDF’s) of the distances between inclusions and the distances between inclusions and the origin of coordinates. The methodology is numerically tested in a light scattering example in which the host particle is eliminated by matching the refractive indices of host particle and medium. The results obtained for this cluster particle are very satisfactory.

1. Introduction
Inhomogeneous particles play an important role in many fields of science like optical particle sizing, astronomy, optical oceanography, photographic science, coatings technology, meteorology and atmospheric science. In particular, particles with inclusions have become an important research topic because they appear in many natural and synthetic systems of scientific and technological interest. For instance, characterization of hollow particles is a subject of interest in chemical engineering, while in atmospheric science the characterization of fog droplets with soot or bacteria inclusions has driven great attention. In climatology the study of water droplets in the atmosphere that contain insoluble inclusions is of essential interest.

Since the beginning of scattering science, the study of the scattering characteristics of particles with inclusions has been an important research topic. The interaction of these particles with radiation, that occurs either naturally, for instance when they are irradiated by sunlight, or artificially, as when they are irradiated by a source in the laboratory, raise a number of interesting technical problems.

The first scattering calculations for particles with inclusions were done for a sphere with a concentric spherical inclusion [1], and have been extended afterward for a concentric multilayered sphere [2]. The exact solution for the light scattering by a spherical host containing one or several non-concentric spherical inclusions has been investigated by several authors [3,4,5,6,7]. The calculation of the scattering characteristics of a particle with inclusions, that is performed numerically, may be very time consuming depending on the complexity of the particle. When the calculations have to be repeated many times as occurs when the model is used to characterize the particle from scattering measurements, the computing load may become unacceptable. For this reason approximated models...
are of great interest in these cases because on one hand they help to reduce the computational time needed to solve them, and in the other hand the solution of the associated inverse problem, needed to characterize the particle, may be simplified.

In this work we have first developed a model for a spherical particle with multiple spherical inclusions in the Rayleigh-Gans (RG) approximation [8]. The RG approximation is valid for particles that present a low optical contrast with respect to the ambient medium, and are also sufficiently small so that the phase shift between incident and scattered light is also small. The conditions of validity for the RG approximation are always fulfilled in X-ray scattering [9] but are more difficult to accomplish in light scattering. Then, based on this approximated model, we have proposed an inverse methodology to estimate the particle parameters. In the proposed methodology, these parameters were: radius of host particle, radius of inclusions and number of inclusions. Finally, a light scattering example is presented.

2. Scattering model of a spherical particle with multiple spherical inclusions in the RG approximation

A group of \( N \) arbitrary particles located at positions given by \( \mathbf{R}_j \) (\( j = 1, \ldots, N \)), scatters light with amplitude electric field, \( E_S \), given by [9]

\[
E_S(\mathbf{R}) = -E_0 \frac{\exp(ikR)}{R} \sum_{j=1}^{N} b_j(q) \exp(-iq\mathbf{R}_j) \tag{1}
\]

where vector \( \mathbf{R} \) (\( |\mathbf{R}| = R \)) indicates the position of the detector, \( E_0 \) is the magnitude of the incident field which in this case is assumed to be polarized perpendicular to the scattering plane, \( q \) is the scattering vector, and \( b_j \) is the scattering length of particle \( j \). Note that time dependence has been omitted.

\[\text{Figure 1. a) Decomposition of particle with inclusions; b) Decomposition of homogeneous particle; and c) Decomposition of particle with inclusions as a sum of spherical particles.}\]

A spherical particle with \( N \) spherical inclusions can be thought as the sum of a spherical particle with holes located where the inclusions are, plus the inclusions, as described in figure 1.a, i.e. \( N_i + 1 \) particles. On the other hand, a homogeneous spherical particle can be decomposed as the sum of a spherical particle with holes, plus inclusions in the same positions as the holes and made of the same material as the homogeneous particle, as shown in figure 1.b. With this in mind, the spherical particle with spherical inclusions can be decomposed as indicated in figure 1.c and in this form represented in...
terms of spherical particles. If the large particle is considered at the origin of coordinates, the amplitude of the electric field can be written as

\[ E_S(\mathbf{R}) = -E_0 \frac{\exp(ikR)}{R} \left[ b_R(\mathbf{q}) + \left( b_r^{(2)}(\mathbf{q}) - b_r^{(1)}(\mathbf{q}) \right) \sum_{j=1}^{N} \exp(-i\mathbf{q} \cdot \mathbf{R}_j) \right], \]

where the scattering lengths of the large sphere and the spherical inclusions are given by:

\[ b_R(\mathbf{q}) = k^2 \left( \frac{n_1 - n_0}{n_0} \right) V_R F_1(q) \quad \text{with} \quad F_1(q) = \left[ \frac{3}{(qR)^3} \sin qR - qR \cos qR \right] \]

\[ b_r^{(i)}(\mathbf{q}) = \frac{k^2}{2\pi} \left( \frac{n_i - n_0}{n_0} \right) V_r F_2(q), \quad i = 1, 2 \quad \text{with} \quad F_2(q) = \left[ \frac{3}{(qR)^3} \sin qr - qr \cos qr \right] \]

Here, \( n_0, n_1 \) and \( n_2 \) are the refractive indices of medium, large particle and inclusions, respectively; \( R \) is the radius of the large particle; \( r \) is the radius of inclusions; \( q \) is the absolute value of the scattering vector \( (q = \frac{4\pi n_0}{\lambda_0} \sin \frac{1}{2} \theta) \); \( k = 2\pi n_0 / \lambda_0 \) is the magnitude of the propagation vector of the incident radiation; \( \theta \) is the scattering angle; and \( \lambda_0 \) is the wavelength of the incident radiation in vacuum.

Thus, the Differential Scattering Cross Section (DSCS) for this particle is given by

\[ \frac{d\sigma(q)}{d\Omega} = \frac{|E_S(\mathbf{R})|^2 R^2}{E_0^2} \left[ b_R(\mathbf{q}) + \left( b_r^{(2)}(\mathbf{q}) - b_r^{(1)}(\mathbf{q}) \right) \sum_{j=1}^{N} \exp(-i\mathbf{q} \cdot \mathbf{R}_j) \right]^2 = \]

\[ \frac{k^4}{4\pi^2 n_0^2} \left( (n_1 - n_0)^2 V_R^2 F_1(q)^2 + \right. \]

\[ \left. (n_2 - n_1)^2 V_r^2 F_2(q)^2 \sum_{j=1}^{N} \sum_{k=1}^{N} \exp[-i\mathbf{q} \cdot (\mathbf{R}_j - \mathbf{R}_k)] \right) + \]

\[ 2(n_1 - n_0)V_R F_1(q)(n_2 - n_1)V_r F_2(q) \sum_{j=1}^{N} \exp(-i\mathbf{q} \cdot \mathbf{R}_j) \}

This formula is valid for a particle with inclusions in fixed positions \( \mathbf{R}_j, j = 1, \ldots, N \). If one considers either a diluted system of identical particles with inclusions whose orientations are randomly distributed in all possible directions, or a particle that is rapidly moving taking all possible orientations, such that the interesting quantity in these two cases is the average value of the DSCS, orientational average, denoted \( \langle \cdot \rangle \), must be taken on the DSCS given before, as follows

\[ \frac{d\sigma(q)}{d\Omega} = \frac{k^4}{4\pi^2 n_0^2} \left( (n_1 - n_0)^2 V_R^2 F_1(q)^2 + \right. \]

\[ \left. (n_2 - n_1)^2 V_r^2 F_2(q)^2 \sum_{j=1}^{N} \sum_{k=1}^{N} \exp[-i\mathbf{q} \cdot (\mathbf{R}_j - \mathbf{R}_k)] \right) + \]

\[ 2(n_1 - n_0)V_R F_1(q)(n_2 - n_1)V_r F_2(q) \sum_{j=1}^{N} \exp(-i\mathbf{q} \cdot \mathbf{R}_j) \}

where the orientational averages can be calculated analytically and are given by

\[ \sum_{j=1}^{N} \sum_{k=1}^{N} \exp[-i\mathbf{q} \cdot (\mathbf{R}_j - \mathbf{R}_k)] = N_i + \sum_{j<k}^{N} \sum_{k=1}^{N} \sin |\mathbf{q}||\Delta \mathbf{R}_{jk}| / |\mathbf{q}| \]

(7)
\[
\sum_{j=1}^{N_i} \exp(-i\mathbf{q} \cdot \mathbf{R}_j) = \sum_{j=1}^{N_i} \frac{\sin |\mathbf{q}||\mathbf{R}_j|}{|\mathbf{q}||\mathbf{R}_j|}
\]

where \( \Delta \mathbf{R}_{jk} = \mathbf{R}_j - \mathbf{R}_k \).

Finally, the orientationally averaged DSCS of a spherical particle with spherical inclusions is given by

\[
\left\langle \frac{d\sigma(q)}{d\Omega} \right\rangle = \frac{k^4}{4\pi^2 n_0^2} \left[ (n_1 - n_0)^2 V_R^2 F_1(q)^2 + N_i (n_2 - n_1)^2 V_r^2 F_2(q)^2 + (n_2 - n_1)^2 V_r^2 \sum_{j} \sum_{k \neq j} \frac{\sin |\mathbf{q}||\Delta \mathbf{R}_{jk}|}{|\mathbf{q}||\Delta \mathbf{R}_{jk}|} + 2(n_1 - n_0)(n_2 - n_1) F_1(q) F_2(q) V_r V_R \sum_{j=1}^{N_i} \frac{\sin |\mathbf{q}||\mathbf{R}_j|}{|\mathbf{q}||\mathbf{R}_j|} \right]
\]

### 3. Inverse problem formulation

In what follows the problem of obtaining the radius of the host particle and the radius and number of inclusions from measurements of the DSCS will be addressed. In this analysis the refractive indices of medium and particles will be considered to be known. The unknown parameters in the model of the DSCS that will be used to fit the measurements are: the radius of host particle, the radius of inclusions, the number of inclusions, the distances of the inclusions to the origin \((N_i)\) and the distances between inclusions \([N_i(N_i-1)/2]\). All these parameters appear non-linearly in the model and totalize \(N_i(N_i + 1)/2 + 3\). A number that, for instance, for \(N_i=20\), amounts to 213. The non-linear form in which the parameters appear and their high number, make the estimation difficult. For that reason it is attractive to explore some other parameterization that could reduce the number of parameters and make most of them to appear linearly.

To accomplish this objective, two discrete Distance Distributions Functions (DDF’s) are defined whose ordinates in vector form are

\[
h = \left[ h_1 \ h_2 \ \ldots \ h_{m_1} \right]^T
\]

\[
f' = \left[ f'_1 \ f'_2 \ \ldots \ f'_{m_2} \right]^T
\]

These ordinates are the number of distances \(r_i\) from the origin of coordinates to the inclusions for \(h\), and the number of distances between inclusions for \(f'\), corresponding to the following distances: \(r_i, i=1, \ldots, m_1\) for \(h\) and \(\Delta r_i, i=1, \ldots, m_2\) for \(f'\). Where \(0 \leq r_i \leq (R-r)\) and \(2r \leq \Delta r_i \leq 2(R-r)\). With these definitions, the sums in (9) can be written as

\[
\sum_{j=1}^{N_i} \frac{\sin |\mathbf{q}||\mathbf{R}_j|}{|\mathbf{q}||\mathbf{R}_j|} = \sum_{j=1}^{m_1} h_j \frac{\sin |\mathbf{q}||r_j|}{|\mathbf{q}||r_j|}
\]

\[
\sum_{j} \sum_{k \neq j} \frac{\sin |\mathbf{q}||\Delta \mathbf{R}_{jk}|}{|\mathbf{q}||\Delta \mathbf{R}_{jk}|} = \sum_{j=1}^{m_1} f_j \frac{\sin |\mathbf{q}||\Delta r_j|}{|\mathbf{q}||\Delta r_j|}
\]

The model of (9) is then written as

\[
\left\langle \frac{d\sigma(q)}{d\Omega} \right\rangle = a(R,q) + b(R,r,q) \sum_{j=1}^{m_1} h_j \frac{\sin |\mathbf{q}||r_j|}{|\mathbf{q}||r_j|} + c(r,q) \left( N_i + \sum_{j=1}^{m_2} f_j \frac{\sin |\mathbf{q}||\Delta r_j|}{|\mathbf{q}||\Delta r_j|} \right)
\]

where \(a(R,q), b(R,r,q)\) and \(c(r,q)\) are easily deduced from (9).

Define now

\[
i_j = \left[ \left\langle \frac{d\sigma(q_1)}{d\Omega} \right\rangle, \left\langle \frac{d\sigma(q_2)}{d\Omega} \right\rangle, \ldots, \left\langle \frac{d\sigma(q_{m_2})}{d\Omega} \right\rangle \right]^T
\]
Then

\[ i_y = a + [BS_1 \quad CS_2] \begin{bmatrix} h \\ f \end{bmatrix} \]  

(16)

with

\[
a = \begin{bmatrix} a(R, q_1) & a(R, q_2) & \ldots & a(R, q_{m_2}) \\ b(R, r, q_1) & 0 & \ldots & 0 \\ b(R, r, q_2) & 0 & \ldots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ b(R, r, q_{m_1}) & 0 & \ldots & 0 \end{bmatrix}^T
\]

(17)

\[
B = \begin{bmatrix} S_{11}^{(1)} & S_{12}^{(1)} & \ldots & S_{1m_1}^{(1)} \\ S_{21}^{(1)} & S_{22}^{(1)} & \ldots & S_{2m_1}^{(1)} \\ \vdots & \vdots & \ddots & \vdots \\ S_{m_1,1}^{(1)} & S_{m_1,2}^{(1)} & \ldots & S_{m_1,m_1}^{(1)} \\ S_{11}^{(2)} & S_{12}^{(2)} & \ldots & S_{1m_2}^{(2)} \\ S_{21}^{(2)} & S_{22}^{(2)} & \ldots & S_{2m_2}^{(2)} \\ \vdots & \vdots & \ddots & \vdots \\ S_{m_2,1}^{(2)} & S_{m_2,2}^{(2)} & \ldots & S_{m_2,m_2}^{(2)} \end{bmatrix}
\]

(18)

\[
S_1 = \begin{bmatrix} c(r, q_1) & 0 & \ldots & 0 \\ 0 & c(r, q_2) & \ldots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \ldots & c(r, q_{m_1}) \end{bmatrix}
\]

(19)

\[
S_2 = \begin{bmatrix} S_{11}^{(2)} & S_{12}^{(2)} & \ldots & S_{1m_2}^{(2)} \\ S_{21}^{(2)} & S_{22}^{(2)} & \ldots & S_{2m_2}^{(2)} \\ \vdots & \vdots & \ddots & \vdots \\ S_{m_2,1}^{(2)} & S_{m_2,2}^{(2)} & \ldots & S_{m_2,m_2}^{(2)} \end{bmatrix}
\]

(20)

and

\[
f = \begin{bmatrix} N_1 & f^T \end{bmatrix}^T
\]

(21)

According to (16), the unknowns are now \( R \) and \( r \), that appear non-linearly in (16), and \( f \) and \( h \), that are in linear fashion. The total number of unknowns is \( m_1 + m_2 + 3 \). Independently of the number of inclusions, this number can be maintained relatively low because \( m_1 \) and \( m_2 \) can be selected rather arbitrarily. Another advantage of this parameterization is that the unknowns are displayed as distributions with the advantage that the correlation between the ordinates of these particular functions simplify the solution of the problem as discussed below.

Assume by the moment that \( r \) and \( R \) are known. Under this assumption, the solution for \( f \) and \( h \) could be obtained in principle by linear least squares. However, the original structure of (16) guarantees that the problem has some degree of ill-conditioning that will require a regularization scheme. If the so called Tikhonov regularization is adopted [10], the following minimization problem must be solved:
In a particular case of a particle with inclusions is analyzed through simulation. Such case arises when in (9) the first and last values of $h$ and $f'$ to be 0, in accordance with what it is expected from both distributions. $\theta_i$ and $\theta_0$ are vectors of zeros of dimensions $m_1$ and $m_2$. $\gamma_1$ and $\gamma_2$ are the regularization parameters that equilibrate the amount of regularization with the amount of fitting. The regularization parameter is selected as in reference [11].

To estimate also $r$ and $R$, the previous scheme is incorporated into the following algorithm which is similar to one reported in reference [11]:

1) Select $q_i$, $i = 1, \ldots, m_1$; $r_i$, $i = 1, \ldots, m_1$; and $\Delta r_i$, $i = 1, \ldots, m_2$.
2) Give a range of possible values for the radii of the inclusions ($r_{\text{min}}$ to $r_{\text{max}}$) and the host sphere ($R_{\text{min}}$ to $R_{\text{max}}$), and select a set of $N_r$ values of $r$ and a set of $N_R$ values of $R$.
3) Calculate $S_i$ and $S_2$.
4) Calculate $a(R)$ for the $N_R$ values of $R$.
5) Calculate $B(r,R)$ for the $N_r \times N_R$ values of $r$ and $R$.
6) Calculate $C(r)$ for the $N_r$ values of $r$.
7) Perform $N_r \times N_R$ estimations of $h$ and $f$ for each one of the selected values of $r$ and $R$.
8) Select as solution of the problem the group ($h, f, r, R$) such that $J(h, f, r, R)$ is minimum.

### 4. Light scattering example

In this example a particular case of a particle with inclusions is analyzed through simulation. Such case arises when in (9) the first and last values of $h$ and $f'$ to be 0, in accordance with what it is expected from both distributions. $\theta_i$ and $\theta_0$ are vectors of zeros of dimensions $m_1$ and $m_2$. $\gamma_1$ and $\gamma_2$ are the regularization parameters that equilibrate the amount of regularization with the amount of fitting. The regularization parameter is selected as in reference [11].

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forming the cluster are assumed to be randomly distributed inside a hypothetical sphere of radius $R = 1 \mu m$. Figure 2 illustrates how the particular configuration of the cluster used in this example looks.

With this information a synthetic “measured” DSCS spectrum is generated using (9) at 180 equally spaced points for most of the whole angular range (1º-180º). The particle cluster is assumed to be illuminated with light of $\lambda_0 = 0.6328 \mu m$ which is polarized perpendicularly to the scattering plane. In order to make the simulations more realistic, the generated spectrum is corrupted with uniformly distributed zero mean random noise of maximum value equal to 5% of the value of the uncorrupted spectrum at each angle. In figure 3 the noisy spectrum versus the scattering angle is shown in full line.

In figure 4.a the exact DDF corresponding to the distances between particles in the cluster is illustrated. In this plot the height of each stem represents the number of particles for each of the real distances existing between the centers of the particles. Because these distances are all different, the resulting stem plot has the shape of a box of height one and width equal to the difference between the larger and the smaller existing distances. This plot represents the particle exact DDF on a non-equally spaced distance coordinate axis. Thus, the information of the distances between the particles is given in a form that is not useful to contrast with the estimated particle DDF which is represented on an equally spaced distance axis as explained in section 3. For that reason the exact DDF of figure 4.a is transformed into one in which the distances are equally spaced. This is done by counting the existing particles in a given distance range and then assigning the resulting number to a distance that is in the middle of that range. The resulting plot is shown in figure 4.b for 30 distances that range between 0.1$\mu m$ and 2$\mu m$, resulting in a step of 0.0655$\mu m$.
With the assumption that the refractive indices of medium and cluster component particles are known, the estimation of \( r, N_i \) and \( f' \) is performed using the method proposed in the previous section. In this example the estimation involves only the DDF between particles, \( f' \), and for that reason the problem is simplified. For the estimation, a distance distribution function with \( m_2 = 100 \) points over an equally spaced distance coordinate ranging from \( \Delta r_1 = 0.02\mu m \) and \( \Delta r_{m2} = 3.38\mu m \) is used. The distance step in this case is 0.034\( \mu m \). In order to compare the distribution of figure 4.b, that represents an equally spaced approximation of the “real” distribution (figure 4.a) with the estimated one, the estimated DDF is normalized by multiplying it by the ratio between the distance step taken for the approximation to the “real” DDF and the distance step of the estimated DDF, i.e. \( 0.0655/0.034 = 1.93 \).

The estimation problem is solved for \( f \) repeatedly with a fixed value of \( \gamma = 1e-06 \) and for values of \( r \) that cover a range between 10% less than its real value (0.2\( \mu m \)) and 10% more than that value, i.e. from 0.18\( \mu m \) and 0.22\( \mu m \). A plot of \( J(f, r) \) vs \( r \) displaying a minimum at \( r = 0.2014 \), is shown in figure 6. In figure 5 the normalized estimated DDF for this value of \( r \) is plotted together with the approximation to the “real” DDF. In figure 3 the estimated DSCS spectrum corresponding to the solution of the inverse problem is shown in dotted line together with the measured spectrum as mentioned before.

**Figure 4.** Distribution function of the distances between particles in the cluster for the light scattering example: a) exact; b) approximated.

**Figure 5.** Distribution function of the distances between particles in the cluster for the light scattering example: real distribution, bar plot; and estimated distribution, full line plot.

**Figure 6.** Functional of (23) as a function of the size of the particles that conform the cluster.
Finally the estimated element $m_2+1$ of $f$ is given by $N_i = 19.30$. As seen, all the estimated values agree within a very small error with the real values and the shape of the distance distribution resembles quite well that of the approximated “real” DDF. From the estimated DDF also a value of $N_i$ can be estimated using the relationship between the number of inclusions and the sum of the DDF ordinates, i.e. $\sum_{i=1}^{m_i} f_i = \frac{N(N_i-1)}{2}$. With this equation a value of $N_i = 19.81$ is obtained, which is even better than the one estimated directly.

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