Stochastic Geometry for Electromagnetic Scattering Modeling

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Abstract: The relationship between statistical distributions of geometrical parameters and scattering properties of particles has been underlined, particularly for optically soft particles. As the calculation of these distributions may be difficult, some simple rules for estimating them are presented in this paper. The method has been validated by the calculation of the scattering properties of a capsule shaped particle.

Keywords: electromagnetic scattering, approximated method, stochastic geometry, complex morphology

1. Introduction

Electromagnetic scattering by particles consisting in a homogeneous material has been studied a lot by numerically solving the Maxwell equations. Solutions depend on the morphology of the particles; this is revealed at the boundary conditions, i.e. the values of the physical quantities on the particle surface. In the case of a homogeneous material a relationship between the scattering properties, the optical indices of the material and the surface characteristics may be expected. The exact calculation of any shaped particle is possible by numerical methods [1]. However, the whole analysis needs a too large computational time if these exact methods are used for the calculation of the optical properties. Actually, the need of fast calculations is important for industrial applications, e.g. particle sizing and particle shape determination by means of optical methods, even if the accuracy is lower. Fitting the accuracy of the calculation with the accuracy of the measurement is sufficient. Thus simple and accurate expressions for the optical properties of particles or crystals are useful to solve inverse problems coming from such optical particle sizing techniques.

Moreover the use of relevant descriptors for the particle geometry can simplify the correspondence between optical and geometrical properties. In the past some optical approximations have been developed for the calculations for optically soft materials [2]. They include relations between the optical properties and the statistical distributions of geometrical parameters [3, 4]. Unfortunately, it appears that the analytical calculation of the statistical distributions can be very tedious even for simply shaped particles. The aim of this paper is to extract from the dedicated literature any information to evaluate the statistical distributions of any convex body and then calculate the scattering properties. The study of non convex particles, e.g. clusters, will be out of the scope of this paper.

2. Relation between scattering properties and statistical distributions

The optical properties of a body depend on the relative refractive index *m*, i.e. the ratio between the refractive indices of the particle and the surrounding medium. Some scatterers dispersed in a liquid are characterized by a low optical contrast, i.e. $|m-1| \ll 1$. In the past, two important analytical approximations [2] have been proposed for such particles: The Van de Hulst approximation, also known as anomalous diffraction (AD) approximation, for large particles and the Rayleigh-Debye-Gans

(RDG) approximation for small particles. From a geometrical point of view, the AD approximation explicitly contains the $D_l(l)$ chord length distribution (CLD) of the scatterer. The chord length l is the distance between two points located on the body surface. RDG approximation considers the $D_P(r)$ distance distribution (DD) of scattering elements inside the scatterer. The distance r is the length between two any points inside the body. Now, we will focus our attention about the scattering of light by small and optically soft particles.

Within the RDG approximation, the differential scattering cross section per unit incident intensity (unpolarized light) is related to the amplitude form factor R. Considering that the object (scatterer) can randomly orientate, this optical property is thus obtained after calculating an average over all the object orientations (Eq.1):

$$\left\langle C_{diff\ sca} \right\rangle = \frac{k^4 V^2}{4\pi^2} |m-1|^2 \left\langle |R|^2 \right\rangle \frac{1+\cos^2\theta}{2} \tag{1}$$

with

$$\left\langle |R|^2 \right\rangle = \int_0^{l_{\max}} \frac{\sin(k_s r)}{k_s r} D_P(r)\, dr \tag{2}$$

$\vec{k}$, θ and $\vec{k}_s$ ($k_s = 2k\sin(\theta/2)$) are the incident wave vector, the scattering angle and the scattering vector, respectively. V is the object volume.

Following the theory of stochastic geometry [3], there exists a relation between the two normalized distributions D_P and D_l for a convex particle:

$$D_P(r) = 12r^2 \int_r^{l_{\max}} D_l(l)(l-r)\, dl / \overline{l^4} \tag{3}$$

l_{max} is the maximum chord length of the particle.

By introducing Eq.3 into Eq.2, one express $\left\langle |R|^2 \right\rangle$ and the differential scattering cross section as a function of the CLD:

$$\left\langle |R|^2 \right\rangle = \frac{12}{\overline{l^4} k_s^4} \left[2 - \int_0^{l_{\max}} \left(k_s l \sin(k_s l) + 2\cos(k_s l) \right) D_l(l)\, dl \right] \tag{4}$$

Eq. 4 shows that D_l is the relevant geometrical characteristic for the scattering cross section of optically soft particles. Equation (4) replaces the equation defining $\left\langle |R|^2 \right\rangle$ that is basically a sextuple integral. As a consequence, the use of CLD presents two advantages: simpler calculations and clear relationship between the particle morphology and the scattering properties.

Two ways can be used for calculating the CLD of a convex body: analytical calculation for the simplest shaped particles or Monte-Carlo Simulations (MCS). The MCS runs are carried out by generating unbiased random lines that cross the particle. The distance between the two points that intersect the body is calculated and recorded. The MC sampling distribution may be visually represented as the discrete probability histogram.

3. Mathematical properties of CLD

Statistical distributions have already been introduced by researchers in the field of the small angle scattering using X-rays (SAXS). Therefore we may compile the main results of the literature especially these ones concerning the SAXS theory. Moreover, additional properties have been discovered by the mathematicians.

We may consider the CLD of any convex body as the CLD of an equivalent spheroid or ellipsoid, i.e. a body with a smooth shape, modified by specific geometrical features. The latter ones are important for the shape of the CLD curve. They consist

in flat faces as crystal facets, parallel (flat or curved) surfaces, parallel tangent planes, edges and corners. They correspond to discontinuities of the distribution density or its derivative.

We begin by investigating the effect of the curvature on CLD. For a convex body with a smooth surface the series expansion of CLD around *l=0* does not contain even terms. The first order term is an explicit function of the mean principal curvatures of the body [5, 6].

Chords intersect the surface at two points in a convex body; some chords are perpendicular to the tangent planes at the two end points and thus the latter ones are parallel. These chords are termed extremal chords. For instance, spheroids have two extremal chords: one for *l=2a* and the other for *l=2b*, *a* and *b* being the semi-axes. A discontinuity of $dD_l(l)/dl$ will occur at these values. Wu and Schmidt [7] have investigated the properties of $D_l(l)$ when the chord is in the neighbourhood of an extremal chord. They give expressions for $D_l(l)$ around the extremal chord values denoted *L*. They show that $D_l(l)$ is continuous for *l=L* whereas $dD_l(l)/dl$ is not.

Ciccariello [8-10] generalized the work of Wu and Schmidt: he considers the property of parallelism between some parts of body boundaries. He studied the case where the locus of the extremal chord ends is a surface. For instance, this surface is a sphere for *l=2R* if the particle is a sphere with radius *R*. One can show that $D_l(l)$ becomes discontinuous for this chord length value. If the parallelism occurs between two partial surfaces of the body, a discontinuity of $D_l(l)$ occurs at *l=L*, *L* being the distance between the two parallel surfaces. The contribution of the parallelism to $D_l(l)$ for $l \to L$ is given by Ciccariello.

The presence of edges leads to additional terms for $D_l(l)$ at *l=0*. Ciccariello et al. [9, 12] and Sobry et al. [11, 13] have shown that $D_l(0)$ is a simple function of the dihedral angle and of the edge length. All the edges contribute to $D_l(0)$. Edges (and corners) also contribute to $dD_l(l)/dl\big|_{l=0}$ [14]. However, it seems difficult to systemize this contribution.

4. Canonical expression for CLD of convex particle

4.1. Principles

Singularities on the surface of a convex body play an important role about the CLD. They correspond to discontinuities of the distribution density and its derivatives. As seen in §3, the CLD discontinuities are slightly proportional to the extension (length or area) of the corresponding singularities. Each singularity is characterized by the values of the chord length and of the density (or its derivative) discontinuity. Generally, the contributions of these singularities are independent of each other and additive. The CLD of particles having a simple shape is not monotonous and show some features as discontinuities as well.

Moreover the integral geometry theory shows that the first order and fourth order CLD moments are related to the surface area *S* and the volume of the body:

$$\bar{l} = 4V/S \quad (5)$$

$$\overline{l^4} = 12V^2/\pi S \quad (6)$$

Then we may establish the following rules for constructing the CLD:

- list the singularities (pairs of parallel tangent planes, pairs of parallel surfaces, edges)
- sort the singularities by increasing chord length values; quantify the corresponding discontinuity of density or of its derivative

- interpolate the density between two singularities by means of polynomial functions
- write the density $D_l(l)$ as a piecewise function
- use the normalization condition for $D_l(l)$ and equations 5-6 for calculating unknown polynomial coefficients in $D_l(l)$.

4.2. Illustration

One considers a capsule shaped particle. This one consists in two hemispheres with radius *R* and a circular cylinder with a radius *R* and length *H*. The capsule is a convex particle with one *l*-discontinuity for *l=2R* due to pairs of parallel surfaces and one $dD_l(l)/dl$-discontinuity for *l=2R+H* due to one pair of parallel tangent planes. Following §4.1 we divide the *l*-range [*0; 2R+H*] into several parts:

- $\left[0; 2R-\Delta l^-\right]$ $D_1(l) = \left[\left(R+3H/16\right)/\left(R^2\left(2R+H\right)\right)\right] l$ see [5-6]
- $\left[2R-\Delta l^-; 2R\right]$ $D_2(l) = D_1\left(2R-\Delta l^-\right) + D_H(l) - D_H\left(2R-\Delta l^-\right)$

with $D_H(l) = -\frac{2H}{S}\ln\left|1-\frac{l}{2R}\right|$ see [8-10]

- $\left[2R; 2R+\Delta l^+\right]$ $D_3(l) = D_4\left(2R+\Delta l^+\right) + D_H(l) - D_H\left(2R+\Delta l^+\right)$
- $\left[2R+\Delta l^+; 2R+H\right]$ $D_4(l) = F_D(l) + a_2\left(2R+H-l\right)^2 + a_3\left(2R+H-l\right)^3$

With $F_D(l) = \frac{4R\left(2R+H-l\right)}{H\left(2R+H\right)^2}$ see [7]

$l = 2R-\Delta l^-$ and $l = 2R+\Delta l^+$ are the length values for which the continuity of CLD function and its derivative is verified respectively :

$D_1\left(2R-\Delta l^-\right) = D_2\left(2R-\Delta l^-\right)$ $\quad dD_1/dl = dD_2/dl$

$D_3\left(2R+\Delta l^+\right) = D_4\left(2R+\Delta l^+\right)$ $\quad dD_3/dl = dD_4/dl$

a_2 and a_3 are determined by using the normalization condition for $D_l(l)$ and the equation 5 for the mean chord length value.

Figure 1 compares the CLD's issued from MCS and calculated by using the previous method. The agreement is good except the CLD part within the $\left[2R+\Delta l^+; 2R+H\right]$ range. In order to check the consequences of this deviation on the optical properties, we calculate the factor $\left\langle |R|^2 \right\rangle$ following Eq.4 for a capsule shaped particle with *R*=0.2µm and *H*=0.4µm. Figure 2 represents the comparison between the $\left\langle |R|^2 \right\rangle$ factors using the two CLD calculations. The slight discrepancy between the two calculations of CLD does not lead to an significant deviation for $\left\langle |R|^2 \right\rangle$.

6. Conclusion

The methodology presented in this paper leads easily to an expression of the CLD that is needed for the scattering properties calculations. Improvements can be done by considering the interactions between singularities if any. This will be studied in the future.

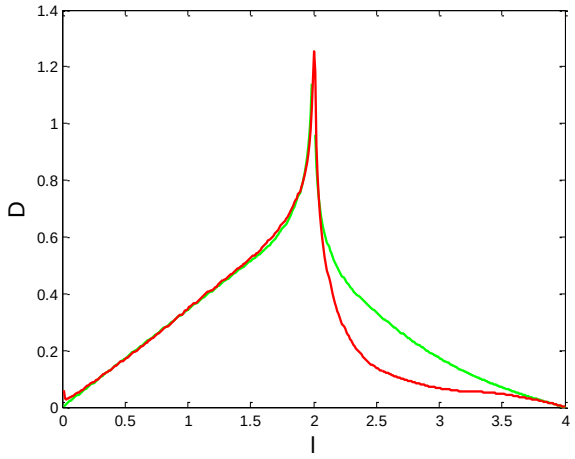

Figure 1: CLD for a capsule (hemisphere radius = 1, cylinder length = 2). red: MCS; green: our method.

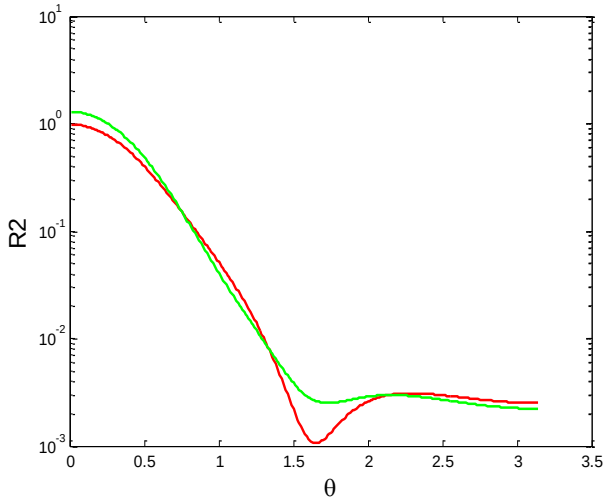

Figure 2: $\left\langle |R|^2 \right\rangle$ as a function of the scattering angle (rad) for a capsule (R = 0.2µm, H = 0.4µm). red: $D_l(l)$ from MCS; green: $D_l(l)$ from our method. λ=450nm.

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