Metrological characterization of micro particles by direct simulation Monte Carlo

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Abstract

Knowledge of the individual micro particle's properties is essential for many applications: medicine and pharmacology (ceramic implants, drug delivery, etc.); micro technologies (polymer, ceramic and composite materials micro manufacture); environment developments. Our study demonstrates the potential of the numerical simulation of single light scattering as a characterization and metrological tool for different types of micro particles. Direct Simulation Monte Carlo (DSMC) model for 3-dimensional single scattering of natural light by randomly oriented suspended homogeneous and isotropic particles with different degree of transparency is presented. The macro shapes can vary from sphere to perfect cube including cube with rounded edges. The micro shape is characterized by roughness in varying degrees. The computer model is validated by comparison with experimental results. Great number of numerical simulations is done and a data base containing polarization phase functions of particles with various size, shape and optical properties is created. The presented numerical technique is applicable to characterization and identification of microparticles of powder materials both of technological (meals, cements, aerosols, drugs, etc.) and natural (terrestrial and atmospheric dust, atmospheric aerosols, cometary’s dust, etc.) origin. The obtained qualitative and quantitative results could be used for the testing of new measurement instruments. The developed DSMC model could be included into integrated computer systems for particle characterization also.

Keywords: powder materials, micro particle metrology, material micro manufacture, micro medicine, light scattering, direct simulation Monte Carlo

1. Introduction

Micro particles are an important component in micro product research, development and quality control of particulate materials, micro technology and material micro manufacture. By this reason metrological characterization and identification of micro particles is an object of continuous interest. At the same time, due to the progress of modern technologies (lasers, computers, etc.), the methods for particle characterisation have changed considerably. Recently, the conventionally used methods like sieve and sedimentation analysis have been displaced by non-invasive methods. The light scattering methods became a major branch of modern particle characterization technology. A good summary of all major particle characterization methods and sample preparation techniques used in particle characterization can be found in [1].

Modeling of the light scattering process, governed by Maxwell's equations, is a non-trivial problem, especially in the case of non-spherical particles. As the analytical methods (classical Lorenz-Mie theory and some extensions) for solving the electromagnetic scattering problem are restricted to some special cases (sphere and spheroid), several semi-analytical and numerical methods are available to deal with more complex shapes of the scattering particles. Light scattering theories and some computational aspects have been reviewed and discussed in [2,3].

We apply Monte Carlo ray tracing as a flexible and powerful tool for simulation of light scattering in geometric optics approximation. The main advantage of this method is the relative simplicity and capability to deal with complex distributed light sources of different types as well as with an ensemble of scattering particles with various shapes and optical properties. It has been applied for spheres [4] and perfect cubes [5]. However, the most of the real particles have non-perfect micro and macro shapes (see Fig.1a). In this study we investigate particles which shape could be approximated by rounded rough cube.

![Fig. 1. (a) Scanning Electron Microphotography of real crystals of KBr with dimensions between 200 µm and 300 µm. (b) Rounded cube.](image)

The main purpose of this paper is to demonstrate the potential of Direct Simulation Monte Carlo of single light scattering as a characterization and metrological tool for different types of micro particles.

2. Computational model

2.1. Physical model

We consider single light scattering by randomly oriented suspended homogeneous and isotropic particles with sizes much larger than the wavelength. The last assumption allows the laws of geometric optics to be applied. The micro particles could be absorbing or non-absorbing. The surrounding media is supposed to be non-absorbing. A laser non–polarized source is assumed. The geometry between illumination and particles is shown on Figure 2.
The light – particle surface interaction is governed by Fresnel law (see Eq. 1-4).

\[
\begin{align*}
T_p &= \frac{2n_1 \cos(\theta_i)}{n_2 \cos(\theta_i) + n_1 \cos(\theta_i)} A_p \tag{1} \\
T_s &= \frac{2n_1 \cos(\theta_i)}{n_1 \cos(\theta_i) + n_2 \cos(\theta_i)} A_s \tag{2} \\
R_p &= \frac{n_2 \cos(\theta_i) - n_1 \cos(\theta_i)}{n_2 \cos(\theta_i) + n_1 \cos(\theta_i)} A_p \tag{3} \\
R_s &= \frac{n_1 \cos(\theta_i) - n_2 \cos(\theta_i)}{n_1 \cos(\theta_i) + n_2 \cos(\theta_i)} A_s \tag{4}
\end{align*}
\]

In these equations \( A_p \) and \( A_s \) represent the amplitudes of parallel and perpendicular components of the electric vector of the incident light with respect to the scattering plane. \( T_p \) (\( R_p \)) and \( T_s \) (\( R_s \)) are the parallel and perpendicular components of the electric vector of the transmitted (reflected) wave respectively. \( n_1 \) (\( n_2 \)) is the refractive index of the first (second) media. \( \theta_i \) (\( \theta_i \)) is the angle between incident (transmitted) ray and the normal to the surface at the point of interaction.

If elliptic polarization arises as a result of total internal reflection (\( n_1 > n_2 \) and \( \theta_i > \theta_{cr} \)),

\[
\sin(\theta_{cr}) = \frac{n_2}{n_1} = n
\]

Fresnel formulas give:

\[
\begin{align*}
|R_p| &= |A_p| \\
|R_s| &= |A_s| \\
\frac{\delta_p}{2} &= -\frac{\sqrt{\sin^2(\theta_i) - n_2^2}}{n_2^2 \cos(\theta_i)} \\
\frac{\delta_s}{2} &= -\frac{\sqrt{\sin^2(\theta_i) - n_1^2}}{n_1^2 \cos(\theta_i)}
\end{align*}
\]

Here \( \delta_p \) (\( \delta_s \)) is the phase jump of the parallel (perpendicular) component of the electrical vector on the optical interface.

2.2. Particle shape

We consider a special class of particle shapes, which vary from a perfect cube to a sphere including essentially rounded cubes. The rounded cube is defined as a section of cube with side \( a \) and sphere with diameter \( d \) which centers coincident (see Fig. 1b). The class of rounded cubes is defined by Eq. (9) for \( k \in [\frac{\sqrt{2}}{2}, 1] \). The parameter \( k \) is called degree of roundness.

\[
k = \frac{a}{d}
\]

The smallest value of the degree of roundness \( \left( k = \frac{\sqrt{2}}{2} \right) \) corresponds to a perfect cube and the biggest one \( (k = 1) \) corresponds to a perfect sphere. The values of \( k \) which belong to the open interval define essentially rounded cubes.

2.3. DSMC scheme

The DSMC algorithm is based on the tracing of photon path for a great number of photons in correspondence with geometric optics laws. As a result of each photon-particle interaction both of the following chains of elementary events are possible: an external reflection; a refraction (the photon gets into the particle), followed by one or more internal (or total internal plus internal) reflections and by a refraction (the photon gets out of the particle). For each of these photon-particle surface interactions Snell law determines the possible propagation direction and the changes of intensities of light polarized parallel and orthogonal to the scattering plane are calculated by means of Fresnel law. At the same time the intensities are used as probabilities for the choice of the outcome of the interaction. The outgoing photon is referred to the corresponding phase angle. The weighted outcomes of tracing of all launched photons and all particle orientations are summed and the final result for the degree of linear polarization \( P_l \) defined by Eq. (10) is
obtained.

\[ P_l = \frac{I_s - I_p}{I_s + I_p} \] (10)

Here \( I_s \) and \( I_p \) are the intensities (squares of the corresponding amplitude modules) of light polarized orthogonal and parallel to the scattering plane. More detailed description of the DSMC algorithm for light scattering by cubic particles can be found in our previous paper [5].

A special shape generator is applied. Three Cartesian frames are used: an absolute coordinate frame for description of incident electric vectors in which the \( x \)-axis is chosen to be parallel to the propagation direction; a local coordinate system which origin coincides with the rounded cube center and the axes are perpendicular to the cube faces (a random particle orientation is achieved by generation of random frame axes directions); another local frame with origin at the interface point for description of the electrical vector changes due to photon-particle interaction. All computations are performed in the local frames and then are transformed in the absolute frame.

3. Numerical simulations

A series of numerical simulations by the presented computer model is performed. All computations are done under the assumptions given in the model description. The number of launched photons in the numerical experiments varies from \( 5 \times 10^6 \) to \( 10^7 \). The refractive index of the surrounding media is taken to be unity.

![Figure 3](image1.png)

**Fig. 3.** Dependence of the degree of linear polarization on the roundness parameter \( k \).

### 3.1. Effect of the shape

At first the proposed computer model is applied to examine the influence of the particle shape on the polarization phase function. Figure 3 shows the degree of linear polarization as a function of the phase angle for five different values of the roundness parameter \( k \). A perfect cube and sphere are included as limit cases of the class of rounded cubes. The refractive index of the scattering particles is 1.54.

The following dependencies become clear from this numerical experiment:
- the results for a perfect cube and a sphere are the same as these obtained by different computer models [5] and [4], respectively. This is a good test for the current model;
- the peaks of the linear polarization function become bigger and move to the backscattering direction with the increase of the roundness degree;
- negative values of the linear polarization function due to backscattering are typical for perfect cubes and shapes with small roundness degrees; the modules of these values decrease when the degree of roundness increases.

### 3.2. Effect of the refractive index

In order to study the qualitative and quantitative behaviour of the polarization phase function depending on the refractive index a data base of great number of curves is created both for absorbing and non-absorbing microparticles with various sizes.

#### 3.2.1. Non-absorbing micro particles

Some of the results from the numerical simulations of light scattering by randomly oriented cubic micro particles with real refractive index are shown on Figure 4. The dependence of degree of linear polarization on the real refractive index for rounded cube is presented in [6].

![Figure 4](image2.png)

**Fig. 4.** Dependence of the degree of linear polarization on the real part of the refractive index.

#### 3.2.2. Absorbing micro particles

Finally, we examine the influence of the absorption on the polarization properties. The computer model is applied for the numerical simulation of light scattering by randomly oriented cubic particles of KBr at wavelength of 0.63 µm. The size of the particles is 100 µm. The real part of the refractive index is 1.5567. The dependence of the degree of linear polarization on the imaginary part of the refractive index is shown on Figure 5.

![Figure 5](image3.png)

**Fig. 5.** Dependence of the degree of linear polarization on the imaginary part of the refractive index.

4. Experimental and numerical study of light scattering by micro crystals of KBr

To measure the polarization phase function of
various types of solid particles a special instrument (see Fig. 2) has been designed in the frame of PROGRA2 funded by CNES. A detail description of the instrument is presented in [7]. Measurements are done in microgravity conditions.

An ensemble of arbitrary oriented crystals of KBr (see Fig. 6) is illuminated by HeNe laser. The sizes of the particles vary from 27.5 µm to 552.5µm. The measured results for the degree of linear polarization are compared with numerical results obtained by the presented DSMC model. The shapes of the particles are approximated by rounded cubes (75%) and parallelepipeds (25%) with uniform distribution of the roundness parameter in the interval $\left[\frac{\sqrt{2}}{2}, 0.72\right]$. The roughness of the particle surface is taken into account by randomly orientation of the scattering plane for 45% of the incident rays. Real part of the refractive index is 1.5567. The imaginary part of the refractive index is used as a free parameter to obtain the best fit for polarization and a value of 0.000075 is found.

The comparison done shows good agreement between experimental and numerical data (see Fig.7). Certain differences between the calculated and measured data are occurred due to the impossibility to approximate more accurate the quite irregular micro shape of the scattering particles.

The comparison done shows good agreement between experimental and numerical data (see Fig.7). Certain differences between the calculated and measured data are occurred due to the impossibility to approximate more accurate the quite irregular micro shape of the scattering particles.

- the polarization phase function on the particle shape;
- the polarization phase function on the real and imaginary part of the refractive index of the scattering particles.

A database containing polarization curves for great variety of shapes, sizes and optical properties of the scattering particles is created.

Experimental and numerical study of light scattering by micro particles of KBr is performed. Computer model is validated by comparison with experimental data.

The developed numerical technique gives powerful tool for metrological characterization and identification of micro particles of powder materials for medical, technological, etc. purposes. The obtained qualitative and quantitative results could be used for designing and testing of new particle characterization instruments.

The computer model could be included into integrated computer systems for particle characterization also.

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**References**